Regional climate response to changes in U.S. aerosol pollution: a review

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Abstract.

Atmospheric aerosols have a direct effect on the planet's radiation balance through both scattering and absorption of solar and terrestrial radiation and they affect the radiation balance indirectly by providing cloud condensation nuclei. Increasingly strict air quality regulations have led to the decline of aerosol precursor emissions over the US throughout the past two decades. Specifically, SO_2 has decreased by 56% between 1980 and 2008 and NO_x decreased by 36% between 1990 and 2008. Recent studies characterizing historical and future radiative forcing from anthropogenic aerosols due to aerosol direct and indirect effects over the US are presented. One study suggests that the net radiative direct forcing from aerosols peaked during the 1980s and 1990s at around –2.5 W m^{-2} and then rapidly declined in magnitude over the next two decades to –1.5 W m^{-2}, following the general trend in the emissions of SO_2 and NO_x; little change (< 0.1 W m^{-2} per decade) in the magnitude of the direct and indirect radiative forcing after 2020 is calculated from future projections of aerosol precursor emissions. The climate response from anthropogenic aerosols is simulated and suggests that regional greenhouse warming, prior to 1980, was entirely masked by a negative radiative forcing from aerosols. Statistically significant changes to the hydrologic cycle and circulation patterns due to aerosols are also observed. Studies suggest air quality improvements have and will likely come at the expense of climate warming, suggesting that future integration of air quality and climate change mitigation policy may be warranted.

1. Introduction.

Aerosols have a direct effect on the Earth's radiative balance through their properties that can scatter and absorb radiation. Aerosols that scatter solar radiation to space have a negative radiative forcing while the opposite is true for aerosols that absorb both solar and terrestrial radiation. Current estimates from the 4th IPCC Assessment Report (AR4) suggest that scattering

alone from anthropogenic aerosols causes an average global radiative forcing of –0.5±0.4 W m^{-2} [*Forster et al.*, 2007]. Aerosols also can increase the availability of both cloud condensation nuclei (CCN) and ice nuclei (IN) that act to seed cloud growth and affect both cloud albedo and cloud lifetime, the latter effects collectively known as the aerosol indirect effect. The net global radiative forcing from the aerosol indirect effect is considerably uncertain, but is estimated to be –0.7±.45 W m^{-2} [*Quaas et al.*, 2009]. The total estimates of the net radiative forcing from anthropogenic aerosols eclipses roughly 1.2 W m^{-2} of the total global radiative forcing (+2.6±0.3 W m^{-2}) from long-lived greenhouse gases (LLGHG) [*Forster et al.*, 2007]. Increasingly strict air quality regulations have raised question as to whether greenhouse gas warming will be exacerbated with a reduction in the magnitude of this –1.2 W m^{-2} cooling term.

Over the past three decades, U.S. air quality management agencies have attempted to mitigate the known health and visibility degradation effects of anthropogenic aerosol by tightening standards on known precursor emissions. As a consequence, emissions from fossil fuel combustion have decreased, sulfur dioxide (SO_2) by 56% between 1980 and 2008 and oxides of nitrogen ($NO_x \equiv NO_2 + NO$) by 36% between 1990 and 2008. Meanwhile, emissions of ammonia (which can contribute to aerosol formation, e.g. ammonium nitrate), black carbon (BC), and organic carbon (OC), have all remained unregulated [*EPA*, 2010]. The reduction of SO_2 and NO_x emissions are of relevance to climate because they can lead to the formation of sulphate and nitrate aerosol, both of which are highly effective at scattering solar radiation. Reductions in sulphate and nitrate aerosol, although beneficial to air quality, may correspondingly decrease the magnitude of the local radiative forcing from anthropogenic aerosols.

Atmospheric particulate pollution causes visibility degradation, is of public health concern, and its relevance to regional climate is of interest in light of decreasing aerosol precursor emissions in the US–a trend that is expected to exacerbate regional greenhouse gas warming [*Brasseur and Roeckner*, 2005; *EPA*, 2010; *Forster et al.*, 2007; *Mickley et al.*, 2012; *Raes and Seinfeld*, 2009; *Ramanathan and Feng*, 2009]. This report investigates and summarizes the current state-of-the-science surrounding the climate response to regional anthropogenic aerosol forcing with a focus on the US.

The mechanisms describing aerosol direct and indirect effects will be presented in section 2. Characterization of the aerosol direct and indirect effect over the US and how regulations have

impacted the aerosol radiative forcing over the US will be presented in section 3, with a focus on a study by Leibensperger et al. [2011a; b]. Results from previous relevant experiments investigating the regional climate response to anthropogenic aerosols over the US will be given in section 4 with considerable attention again given to the Leibensperger et al. study. Finally, section 5 will be a brief conclusion of the presented material.

2. The aerosol direct and indirect effects

2.1 aerosol direct effects

The direct radiative forcing is dependent upon optical factors unique to each aerosol. These include the single scatter albedo, the extinction coefficient, and the scattering phase function – each varying as a function of light wavelength, particle size and the ambient aerosol concentration. The efficiency of scattering and absorption by aerosols is also a strong function of aerosol composition and relative humidity [*Seinfeld and Pandis*, 2006]. The aggregate effects of these factors affecting light extinction result in varying radiative forcings. For example, the net global mean radiative forcing from sulfate, nitrate, and OC aerosol direct effects is estimated to be –0.4 W m^{-2}, –0.1 W m^{-2}, and –0.05 W m^{-2}, respectively [*Forster et al.*, 2007]. The BC component of soot absorbs both infrared and short wave radiation, yielding a positive global mean radiative forcing that modeling experiments believe to be in the range of +0.4 to +1.2 W m^{-2} [*Ramanathan and Carmichael*, 2008]. Additionally, the surface albedo over which aerosols scatter and absorb radiation greatly affects the net top of atmosphere (TOA) forcing from the aerosol direct effect; this interplay can amplify the positive radiative forcing from BC over snow, ice, and clouds [*Flanner et al.*, 2007].

The net radiative forcing from a mixture of primarily scattering (e.g. sulphate) and absorbing (e.g. BC) aerosols is dependent upon how the two materials are mixed throughout the aerosol plume. Aerosols rarely exist as a single species arising from a single source. They are commonly found as a mixture of many chemical components from multiple sources. Two extremes are external and internal mixtures. In external mixture, each aerosol is comprised of one chemical species (e.g. a population of pure NH_4NO_3 particles mixed with pure BC aerosol). In an internal mixture, all aerosols of a particular size contain a uniform mixture of chemical species. These two extremes of aerosol composition have both direct and indirect radiative implications. In an

external mixture, absorbing components will absorb and scattering components will absorb very little, while scattering components will scatter and absorbing components will scatter less. In an internal mixture, if an absorbing component is present, all aerosols will absorb, to some extent. Likewise, in an internal mixture if hygroscopic components are present then each aerosol grows as the relative humidity is increased. Only the hygroscopic particles in an external mixture will grow significantly as RH is increased [*Seinfeld and Pandis*, 2006]. This is particularly relevant in the context of the aerosol indirect effect.

2.2 Aerosol indirect effects

Like the direct effect, the indirect aerosol effect is dependent on the physical and chemical properties of the ambient aerosol concentration, in particular, properties that affect the particle's ability to act as CCN or IN. Such properties include both aerosol diameter and composition as well as the mixing state of the aerosol (internal vs. external mixture). The first indirect aerosol effect, known as the "cloud albedo effect", is a brightening of clouds from an increase in the number of aerosols that form cloud droplets. The increase in number concentration causes a fixed amount of liquid water to be distributed over a greater number of droplets [*Twomey*, 1974]. The second indirect aerosol effect, known as the "cloud lifetime effect", is the change of cloud liquid water content, cloud top height, and cloud lifetime through aerosol effects on CCN and IN [*Albrecht*, 1989].

The IPCC AR4 estimates the current net global radiative forcing from anthropogenic aerosol to be –1.2 W m^{-2} (aerosol direct effect contributing –0.7±.45 W m^{-2} and aerosol indirect effect contributing –0.5±0.4 W m^{-2}) while the total anthropogenic radiative forcing from LLGHGs is around +2.6±0.3 W m^{-2}. Ultimately, the IPCC AR4 and other studies acknowledge that the uncertainties in the magnitude of aerosol direct and indirect effects lead to uncertainties in projections of both future global mean temperatures and climate sensitivity [*Andreae et al.*, 2005; *Forster et al.*, 2007].

3. Characterizing the aerosol direct and indirect radiative forcing over the US

The US has been identified as a unique environment to examine the relationship between aerosol pollution and climate response because of the dramatic changes in aerosol concentrations over half a century that have been attributed to regulatory action (increasing from 1950 to 1990, and

then decreasing from 1990 to 2010) [*EPA*, 2010; *Leibensperger et al.*, 2011b]. Calculating the corresponding changes to the net radiative forcing over the US from 1950 to mid-21st century from aerosol direct and indirect radiative effects was an objective of the Leibensperger et al. [2011b] study.

3.1 Characterizing the radiative forcing over the US due to aerosol direct effect

The Leibensperger et al. study used the Goddard Earth Observing System-Chemistry (GEOS-Chem) Chemical Transport Model (CTM) [*Bey et al.*, 2001] to build both historical and future trends in ambient aerosol mass concentrations over the US of sulfate, nitrate, ammonium, primary and secondary organic aerosol, BC, sea salt and soil dust. The Goddard Institute for Space Studies General Circulation Model 3 (GISS GCM 3) was then used to calculate the corresponding direct and indirect radiative forcings. The radiative forcing calculations for the direct effect were performed for both internal and external mixtures and were calculated by removing each aerosol chemical species from the simulation while keeping other aerosol chemical species unchanged. The radiative forcing from anthropogenic aerosols was calculated by performing a base-case simulation with anthropogenic aerosols and then performing a simulation in which all aerosol components that are of anthropogenic origin are removed. The calculated aerosol direct forcing from contributing US anthropogenic aerosols as a function of decade (1950—2050) are presented in Figure 1. The results in Figure 1 are from the eastern US (east of 100^{o} W) and are from internally mixed aerosol simulations. These results suggest that the net radiative direct forcing from aerosols peaked during the 1980s and 1990s around –2.5 W m^{-2} and then rapidly declined in magnitude over the next two decades to –1.5 W m^{-2}, following the general trend in the emissions of SO_2 and NO_x. Future projections of reduced sulphate, nitrate, BC, and OC mass concentrations from increasingly regulated emissions drive a decrease in the magnitude of the net radiative forcing per decade, but the decreases are marginal after 2020.

3.2 Characterizing the radiative forcing over the US due to aerosol indirect effect

In the Leibensperger et al. study, the cloud albedo effect is quantified through changes to the number concentration of cloud liquid water droplets, N_c, from aerosols acting as CCN. N_c, is calculated from the concentration of water soluble aerosol ions, m_i using the equation given by Boucher and Lohmann [1995]:

$$log\ N_c = A + B\ log\ m_i \quad (1)$$

The size of the effective cloud droplet radius is directly related to N_c through the relation:

$$r_e = \kappa^{\frac{1}{3}} \left[\frac{3L}{4\pi N_c}\right]^{\frac{1}{3}} \quad (2)$$

where L is the liquid water content of the cloud (cm^3 water per cm^3 air), and κ is a constant (0.67 over land, 0.80 over ocean). As N_c increases, the effective cloud droplet radius decreases, causing an increase in the surface area to volume ratio that leads to an increase in the cloud albedo. The both aerosol indirect effects were calculated after the climate simulations were performed ("offline") and does not account for climate feedbacks. The total indirect radiative forcing from eastern US (east of 100° W) anthropogenic aerosols as a function of decade is presented in Figure 2. Like the direct forcing, the magnitude of the total indirect radiative forcing peaks during the 1980s and 1990s around –2 W m^{-2} and then rapidly decreases in magnitude to less than –1.5 W m^{-2} by 2010 as a result of reductions in particulate pollution.

Leibensberger et al. observe that declines in SO_2 emissions decreased the magnitude of the radiative forcing from both aerosol direct and indirect effects over the eastern US from 1990 to 2010 by 1.8 W m^{-2}. The error in all radiative forcing calculations in the Leibensperger et al. study is estimated to be around 50%.

4. Climate response to aerosol direct and indirect radiative forcing over the US

Many global climate modeling studies have examined large-scale aerosol-climate interaction and the associated uncertainties. However, given the short lifetime of aerosols relative to LLGHGs (days to weeks vs. decades to centuries), regional assessments of aerosol-climate interaction are of interest [*Leibensperger et al.*, 2011a; *Schulz et al.*, 2006]. Much attention has been centered on the issue of what, if any, regional climate response is provoked from a given change in radiative forcing that is driven by aerosols. A collection of modeling studies reveal climate responses to aerosol effects of varying magnitudes and directions on regional [*Fischer-Bruns et al.*, 2009; *Jacobson*, 2008; *Mickley et al.*, 2012; *Wang et al.*, 2009] to hemispheric scales [*Levy et al.*, 2008; *Shindell et al.*, 2010].

Using the GISS GCM 3, Mickley et al. [2012] observed a regional increase in annual mean surface temperature of 0.4 to 0.6 K in the eastern US as a result of removing all aerosols over these areas (the total aerosol optical depth over the US was calculated to be 90% anthropogenic in the Mickley et al. study). However, climate response to aerosol forcings are not limited to changes in temperature; aerosol induced strengthening of tropical circulation patterns have been observed in climate modeling studies [*Ming and Ramaswamy*, 2011].

4.1 Surface temperature response to US aerosol loads

Leibensperger et al. [2011a] examined the regional climate response to the changes in anthropogenic aerosols over the US for the historical period of 1950—2010 and the future period of 2010—2050. The change in annual mean surface temperature over the mid-Atlantic due to both the direct and indirect aerosol forcing is presented in Figure 3. The variability in the temperature trend closely matches that of the decadal mean radiative forcing due to US anthropogenic aerosols. The annual mean temperature change from aerosols is between –0.6 and –0.8 ^{o}C and it coincides with the net direct and indirect radiative forcing during the 1980s and 1990s. Using a model simulation that incorporates historical aerosol loads, Leibensperger et al. well reproduce the lack of significant warming that is observed in the GISTEMP record (http://data.giss.nasa.gov/gistemp/) over the mid-Atlantic from 1960—1979 (+0.01 ± 0.20 ^{o}C decade^{-1}). When anthropogenic aerosols are removed over this period, the model, still forced by LLGHG emissions, reveals a significant warming trend (+0.30 ± 0.19 ^{o}C decade^{-1}), thus suggesting that regional greenhouse warming prior to 1980 was entirely masked by a negative radiative forcing from the aerosols. Leibensperger et al. also observe that most of the 0.5 ^{o}C warming that is projected to occur between 1980 and 2050 due to the continued reduction of aerosols occurs by the year 2010 (Figure 4).

Figure 5a shows the observed change in temperature between 1930 and 1990 from the GISTEMP. An area of maximum cooling is observed over a portion of the south central US that has been called the “warming hole” because of the strong cooling trend that is observed coincident with an era of increasing global mean temperature [*Kunkel et al.*, 2006; *Pan et al.*, 2004]. Leibensperger et al., suggest that the origin of the “warming hole” may arise from the effects of aerosol loads over the eastern US (Figure 5b).

Furthermore, Mickley et al. [2012] run a 40-year simulation (2010—2050) of eastern US temperatures following the A1B scenario, both with and without aerosols over the US. Mickley et al. [2012] find that in both scenarios a 9-year running mean reveals that there is around a 0.8 K increase in annual temperatures over the entire simulation period, with a mean difference of 0.5 K between the runs with and without aerosols. Daily mean temperatures during Northeast heat waves in the model were are also amplified by as much as 1-2 K.

4.2 The hydrologic cycle and circulation pattern response to US aerosol loads

Statistically significant changes to the hydrological cycle over the US were additionally attributed to US anthropogenic aerosols in the Leibensperger et al. study. Aerosols can have an effect on how the water vapor is transported and stored in the atmosphere by reducing the amount of available solar energy that ultimately leads to evaporation and through the "cloud lifetime effect". Leibensperger et al. find that annual mean precipitation decreased (0.02—0.20 mm day^{-1}) over the eastern US, but increased (0.02—0.20 mm day^{-1}) over the south central portion of the US between 1970 and 1990 (Figure 6a) – the latter result being attributed to aerosol induced changes in summertime circulation. They conclude that anthropogenic aerosols cooled sea surface temperatures over the North Atlantic causing a strengthening in both the Bermuda High and the onshore flow rate of moist air over the southeastern US that serves as the primary source of summertime water vapor. This change in circulation due to aerosols is thought to magnify the observed cooling over the so-called "warming hole" [*Kunkel et al.*, 2006; *Leibensperger et al.*, 2011a; *Pan et al.*, 2004]. Mickley et al. [2012] find annual average precipitation rates increased (0.1—0.3 mm day^{-1}) over the eastern US when aerosols are removed, consistent with precipitation decreases over the eastern US as a result of 1970—1990 aerosol loads found in Leibensperger et al. [2011a]. However, there is some disagreement in the response of cloud cover to aerosols over the US, although the variety of the simulated clouds presented here are dissimilar. Leibensperger et al. [2011a] propose that the mean annual total cloud cover extent from stratiform clouds increases over both the south central US and the eastern US from anthropogenic aerosol effects (Figure 6b), but Mickley et al. [2012] find increases (0.1-1.0%) in moist convective cloud cover extent when aerosols are removed.

5. Conclusion

Atmospheric aerosols have a direct effect on the planet's radiation balance through scattering and absorption of solar and terrestrial radiation, and they affect the radiation balance indirectly by providing cloud condensation and ice nuclei. Increasingly strict air quality regulations have led to the decline of aerosol precursor emissions over the US throughout the past three decades. Specifically, SO_2 has decreased by 56% between 1980 and 2008 and NO_x decreased by 36% between 1990 and 2008. Studies suggest that these and future air quality improvements have and will likely come at the expense of climate warming, suggesting that future integration of air quality and climate change mitigation policy is warranted.

In a two-part study, Leibensperger et al. [2011a; b] first estimate the total direct and indirect radiative forcing from anthropogenic aerosols over the US from historical and projected emissions. They build aerosol mass concentrations with the Goddard Earth Observing System-Chemistry (GEOS-Chem) Chemical Transport Model and then calculate the resultant radiative forcing from the aerosols with the Goddard Institute for Space Studies General Circulation Model 3 (GISS GCM 3). Their study suggests that the net radiative direct forcing from aerosols peaked during the 1980s and 1990s at around –2.5 W m^{-2} and then rapidly declined in magnitude over the next two decades to –1.5 W m^{-2}, following the general trend in the emissions of SO_2 and NO_x. Future projections show small decreases (< 0.1 W m^{-2} per decade) in the magnitude of the direct and indirect radiative forcing after 2020.

Leibensperger et al. examined the regional climate response to the changes in anthropogenic aerosols over the US. They find that the magnitude of the radiative forcing from aerosols from 1950 to 2010 peaked during the 1980s and 1990s and coincided with the greatest annual mean temperature change (–0.6 to –0.8 ^{o}C) during the same time interval. Leibensperger et al. also hypothesize that aerosol effects may be the cause of the observed "warming hole" over the south central US [*Kunkel et al.*, 2006; *Pan et al.*, 2004].

Leibensperger et al. [2011a; b] and other studies suggest that the climate response to US anthropogenic aerosol loads is vigorous. Current climate change mitigation strategies must account for the direct and indirect effects from anthropogenic aerosols, in addition to long-lived greenhouse gas emissions, if objectives are to prevent potentially dangerous anthropogenic interference with the climate system.

Acknowledgements

D.J. Rasmussen would like to acknowledge Prof. Cappa and two anonymous reviewers for their helpful comments.

6. References

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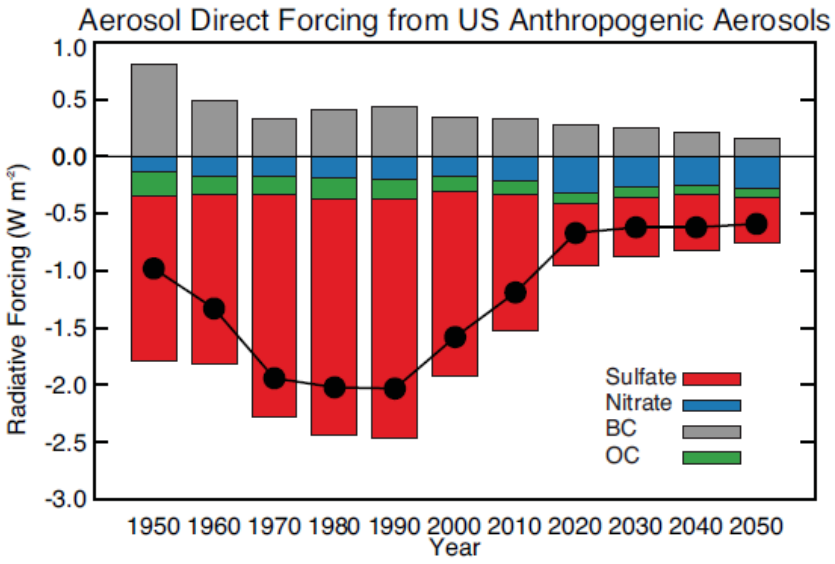
349 **Figures.**



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351 **Figure 1.** From Leibensperger et al. [2011a]: Aerosol direct forcing from eastern US (east of
352 100^{o} W) anthropogenic aerosols as a function of decade. Black circles are the net direct
353 radiative forcing for each decade. Calculations are from an internal aerosol mixture.

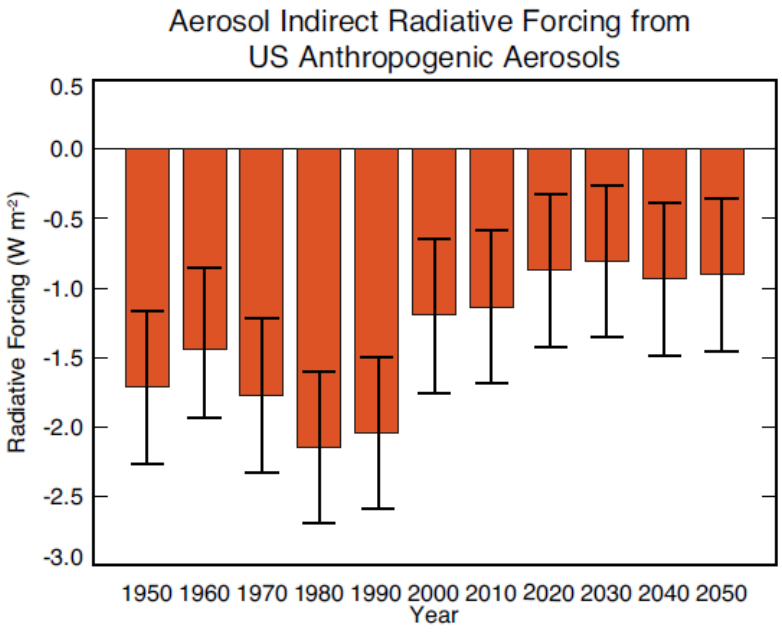

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Figure 2. From Leibensperger et al. [2011a]: The total indirect (cloud albedo + cloud lifetime) radiative effect due to eastern US (east of 100^{o} W) anthropogenic aerosol calculated from the difference of two 50-year climate equilibrium simulations (with anthropogenic aerosols vs. without). The error bars represent the confidence interval at the 95th percentile calculated from inter-annual variability.

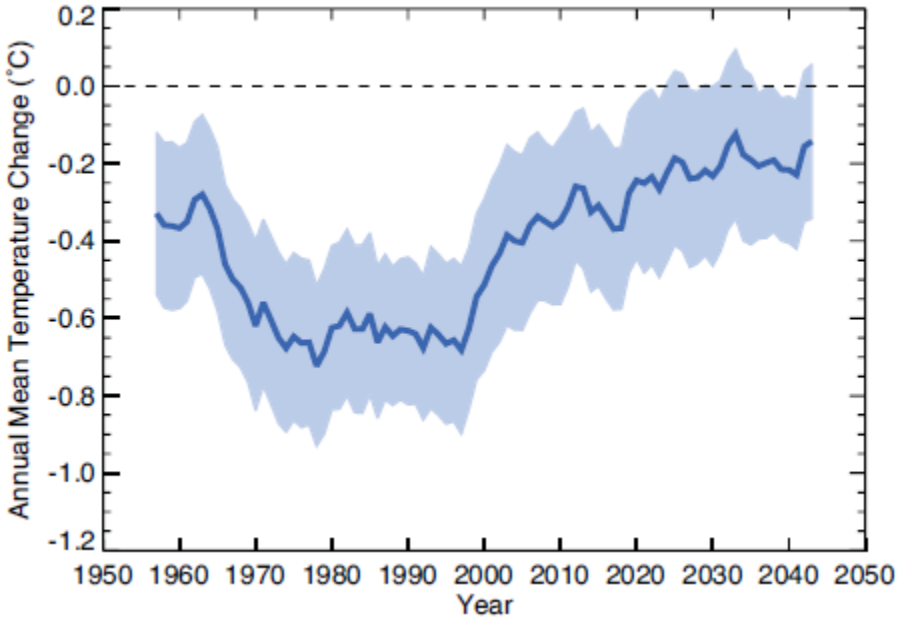
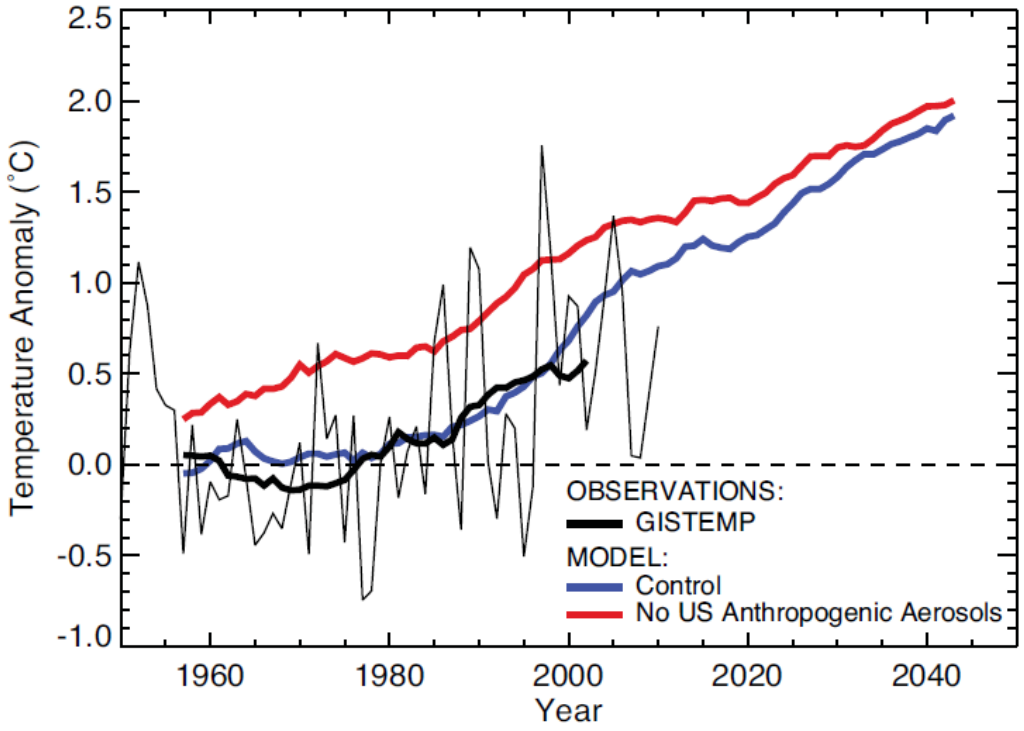


Figure 3. From Leibensperger et al. [2011a]: The change in annual mean surface temperature (^{o}C) over the US mid-Atlantic region attributable to US anthropogenic aerosols calculated by differencing a simulation with radiative forcing from greenhouse gases and aerosols (both indirect and direct effects) and a simulation with zero anthropogenic aerosol. Shading represents the confidence interval at the 95th percentile.



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367 **Figure 4.** From Leibensperger et al. [2011a]: Trends in annual mean surface air temperature
368 (^{o}C) over the mid-Atlantic US from 1950—2050. Observations are the anomaly relative to
369 the 1951—1980 mean and are shown for a 15-year running average (thick black line) and for
370 individual years (thin black line). Model temperatures are from a 5-member ensemble mean
371 and are relative to the 1951—1980 mean of the control simulation. Modeled temperatures are
372 shown as 15-year running averages.

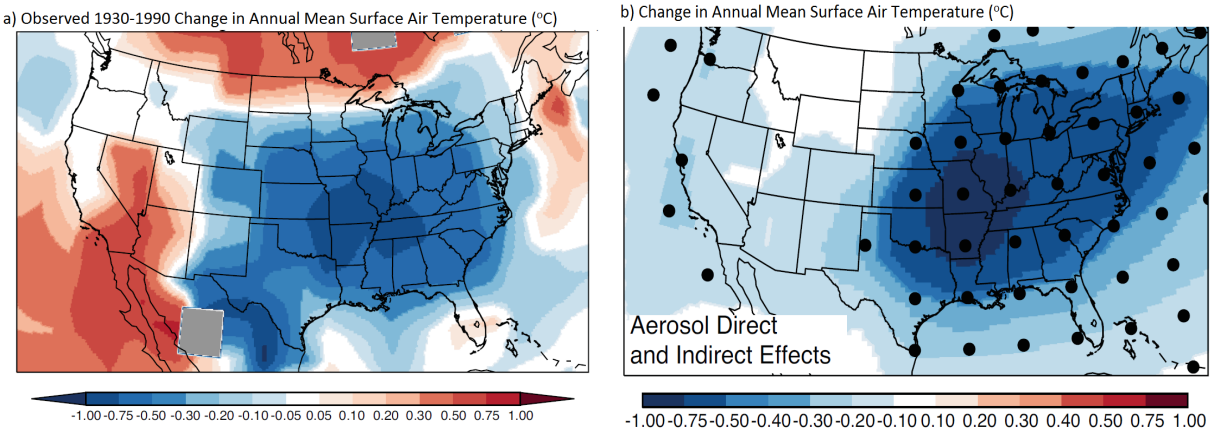

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Figure 5. From Leibensperger et al. [2011a]: a) Observed change in annual mean surface air temperature (^{o}C) between 1930 and 1990. Observations are from the NASA GISS Surface Analysis (GISTEMP) b) The effect of U.S. anthropogenic aerosols on annual mean surface air temperature (^{o}C) for the 1970—1990 period (time of peak 20th century aerosol load). Temperatures are the mean difference between simulations including vs. excluding U.S. anthropogenic aerosols. Black dots are where changes are significant at the 95th percentile.

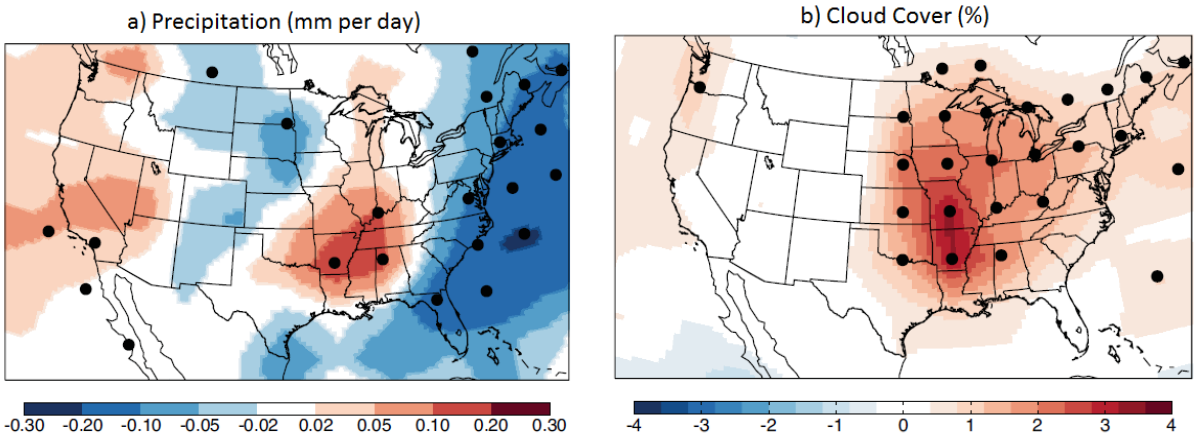

Figure 6. a) From Leibensperger et al. [2011a]: The difference in (a) precipitation (mm day-1) and (b) cloud cover (%) between GCM simulations including US anthropogenic aerosols (both direct and indirect effects) and excluding US anthropogenic aerosols. Black dots are where changes are significant at the 95th percentile.