AEROSOLS

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In Chapter 4, we examined evidence suggesting that human production of aerosol particles has local and regional impacts on clouds, precipitation, and atmospheric temperature. In this chapter we examine the evidence indicating potential impacts of anthropogenic aerosol on global climate.

Estimating the effects of aerosol on climate is particularly challenging. We noted in Chapter~8 that the radiative response to aerosol particles vary with size and chemical composition of the particles relative to the wavelength of the incident radiation. Moreover, because most aerosol particles are heterogeneous in structure, some components of the particles are very absorbing while others reflecting.

Aerosol particles also have limited lifetimes in the atmosphere. Particles greater than a few micrometers may survive for only a few days, while particles on the order of 0.1 μ m and less may reside in the lower troposphere for several weeks. This results in pronounced regional and hemispheric variations in aerosol concentrations.

Unfortunately, there have been few systematic long term observations of aerosol, their size-spectra, and chemical composition. It is, therefore, usually necessary to resort to a variety of less-than-direct measurements, regional field campaigns, and global models to infer changes in aerosol concentrations and their radiative effects.

As discussed in Chapter 4, aerosol particles can have direct effects on atmospheric radiation as well as indirect effects through their impact on cloud microstructure. We shall examine their direct and indirect effects separately below.

Direct Aerosol Effects

It is well known that aerosol particles in polluted urban areas deplete direct solar radiation by about 15\%, sometimes more in winter and less in summer Landsberg, 1970. Less well known, however, is how far the pollution-caused aerosol extend from the urban areas. Schwartz 1989 summarized measurements of the concentrations of sulphate or sulphur aerosol at remote locations in both the northern and southern hemispheres.

These particles form from sulphur dioxide emitted naturally largely by decay of plant and animal matter, by wildland fires, by volcanos, and by anthropogenic activity. The particles form primarily from the in situ oxidation of sulphur dioxide either as a primary gas or as an intermediate stage of oxidation. Two mechanisms for aerosol formation from sulphur dioxide are: 1 dissolving in cloud droplets to create sulfurous acid, which oxides further to form sulphuric acid aerosol particles, and 2 photochemical oxidation to form sulphate particles. Although sulphate particles are not the only human-caused aerosol,

they are certainly prolific, so identification of their distribution is important to understanding the role of human-caused aerosol on climate.

Anthropogenic sulphur dioxide emissions have increased to their present level almost entirely within the last 100 years Cullis and Hirschler, 1980 and, furthermore, as summarized by Schwartz 1989, the bulk of those emissions are in the northern hemisphere. Aerosol sulphate is quite common at remote sites in both the northern and southern hemispheres, with northern hemispheric concentrations substantially exceeding those in the southern hemisphere. At several remote sites the observed high aerosol sulphate concentrations have been attributed to transport from regions of industrial activity over 1000 kilometers away Prahm et al., 1976; Wolff, 1986. There is also evidence that sulphate concentrations have increased substantially over the last century in polar ice at northern hemispheric sites Barrie et al., 1985; Neftel et al., 1985; Mayewski et al., 1986 but there is no such increase in Antarctica Delmas and Boutron, 1980; Herron, 1982. While it is difficult to make accurate estimates of the concentration of sulphate aerosol, and its spatial and temporal variability, there is little doubt that it is increasing, particularly within a few thousand kilometers of industrial regions. As an example, shows the drastic reduction in visibility that has occurred in the eastern United States since 1948. Much of this visibility degradation is attributable to sulfate particles. Black carbon has also been identified as a major forcing aerosol of climate e.g., see Menon et al., 2002; Hansen et al., 2005.

An indirect measure of aerosol concentrations is the electrical conductivity of the air. The electrical conductivity, in turn, is controlled by the concentration and mobility of ions and small, charged aerosol particles. .In general, increases in the concentration of aerosol particles decreases conductivity because the more highly concentrated aerosol particles collect small ions and charged small aerosol particles, thus immobilizing the charge. An exception is radioactive contamination which makes the atmosphere more conductive. Extensive conductivity measurements were made during the first half of this century, but unfortunately the practice has not been continued in the latter part of the century. Cobb and Wells 1970 summarized the results of a few such measurements that were collected from 1907 to 1970. In general, they suggested a 20\% decrease in conductivity took place in the North Atlantic over this period. This corresponds to roughly a doubling of small aerosol particle concentrations over the area. Limited data in the southern Pacific show no such trends.

In general, there is compelling evidence that anthropogenic activity is increasing the concentration of tropospheric aerosol, particularly in the northern hemisphere. Let us now examine the potential impacts of those increased aerosol concentrations on climate. Modeling studies indicate that naturally occurring aerosol particles can affect climate and impact the global circulation of the atmosphere Hansen et al., 1980; Randall et al., 1984; Tanre et al., 1984; Coakley and Cess, 1985; Ramaswamy, 1988; Hansen et al., 1988. The responses of those models to naturally occurring aerosols, varies with the concentration and type of aerosol, the characteristics of the underlying surface i.e., the surface albedo, the solar zenith angle, cloud cover, and the way the models treat ocean responses. The latter effect is particularly important since, if the ocean

temperatures are fixed, the oceans serve as an infinite heat reservoir, and no long-term global temperature responses can be expected Coakley et al., 1987. Furthermore, unless sea surface temperatures can vary, important feedbacks such as variations of the flux of moisture from the ocean cannot occur e.g., Ramanathan, 1981.

In general, absorption of solar radiation by aerosols reduce solar heating at the surface while it heats the layer of air in which the aerosols reside. The impact of aerosols on the surface, however, varies with the albedo of the underlying surface. If a surface has a relatively low albedo such as over the ocean, a given aerosol may increase the surface albedo, while over the higher albedo deserts, it may decrease it. In general, the impact of aerosol on albedo dominates over absorption at most latitudes, but in higher latitudes over snow- or ice-covered surfaces, aerosol absorption can dominate Charlson et al., 1992. Recent work, however, has suggested that the darkening of snow and ice by aerosols particularly black carbon results in a more important aerosol effect Hansen and Nazarenko, 2004.

The atmospheric response to aerosol heating also varies widely depending on the height and depth of the aerosol layer, and on the basic stability of the layer.

While the major forcing of aerosols is regional in nature, there are indications from model experiments that their influence may impact global circulations. Chung and Ramanathan 2003, for example, examined the influence of the south Asian haze on the general circulation. The south Asian haze extends over an area about the size of the United States and covers the south Asian continent to the Arabian Sea and from the bay of Bengal to the Indian Ocean intratropical convergence zone. Chung and Ramanathan 2003 estimate that roughly 75\% of the particles in the haze are emitted by human activities. In their simulations a radiative heating profile was imposed in the lower atmosphere in the NCAR three dimensional global model CCM3 that corresponds to that estimated for the south Asian haze layer. The haze heating was imposed only during the dry season of November to April. In the region of haze heating, weak upward motion develops with corresponding divergence flow in the upper troposphere.

In response to that regional circulation, rainfall is enhanced over the Indian peninsula and suppressed in southwest Asia. Suppressed rainfall over southwest Asia and the western equatorial Pacific is a result of compensating subsidence in the regions outside the regions of dust-enhanced upward motions. Note that the suppressed convection in the tropical western Pacific produces a weaker zonal gradient in latent heating by deep convection which weakens tradewinds, and thereby deepens the ocean thermocline in the eastern Pacific basin. This leads to a weaker zonal gradient of SST which further weakens the tradewinds. Thus the El-Nino Southern Oscillation ENSO circulation field is modulated. Their simulations also suggest that the regional heating by haze perturb the so called Arctic Oscillation Thompson and Wallace, 2000; Thompson et al., 2000 which has been shown to impact northern hemisphere climate.

While these simulations are quite simple, they do indicate that aerosol-induced

regional heating perturbations has the potential of altering circulations and climate over a much larger area. Heating of the air in which the aerosols reside can result in stabilization of a moist moderately stable layer and shut down deep convection and precipitation which could have important climatic implications through the hydrological cycle. In other regions where there is not sufficient moisture or instability to support deep convection, aerosol impacts would be less. This effect of aerosols has been termed the "semidirect" effect Hansen et al. 1997. The reduction in cloud cover associated with this effect can alter the surface energy budget significantly. If the aerosols comprise a large fraction of soot, such as the south Asian haze, then warming in the aerosol layer can nearly totally dessicate stratocumulus cloud layers and alter the properties of the tradewind cumulus layer Ackerman et al., 2000a. General circulation model simulations by Menon et al. 2002 suggest that black carbon emissions over China may be producing changes in the general circulation, which contributes to observed increases in summer flooding in south China and drought in north China. Thus in spite of the fact that anthropogenic aerosol there is regionally concentrated, the potential for global impacts is great.

Another facet of aerosol direct or semi-direct affects is on the nucleation of cloud droplets and thus the concentration of droplets in clouds. Conant et al. 2002 computed the effects of carbon black aerosols on cloud droplet nucleation

Heating parameters of 0.1 and 0.2 are chosen for the two droplet sizes, respectively. The solid curve represents the no-heating case, the dashed curve represents the heating case. Particles are assumed to have the hygroscopic properties of sulfate.illustrates the equilibrium supersaturation over solution drops containing carbon black aerosols. The peak in the curves, called Kohler curves, represents the supersaturation that must be attained in a cloud in order to form growing cloud droplets. If the cooling rate in clouds which is normally proportional to updraft velocity is not large enough to exceed the peak values in the curves, then the aerosol particles of the size indicated cannot form a cloud droplet.

Their calculations showed that since carbonaceous particles are strongly absorbing of solar radiation, the warming of those aerosol particles elevates the peak supersaturation. Thus, for example, if many of aerosols are 0.1 micrometers in diameter, and peak supersaturations are less 0.01%, those particles will not be activated to form cloud droplets owing to absorption of solar radiation. This effect is most pronounced in low supersaturation clouds such as fogs. Moreover, this effect is greatest for larger aerosol particles since the absorption cross section is proportional to the area of the particle. Its main impact is on giant CCN see Nenes et al., 2002 which we have seen in Chapter 4 can initiate or speed up the warm-rain collision and coalescence process. Thus if the carboneous particles absorb solar radiation they may not become big enough to initiate collision and coalescence which, in turn, can act to suppress warm rain processes.

Aerosol Impacts on Clouds--The Twomey Effect

Clouds, we have seen, are good reflectors of solar radiation and therefore contribute significantly to the net albedo of the Earth system. We thus ask, how might aerosol particles originating through anthropogenic activity influence the radiative properties of clouds and thereby affect climate?

First of all, there are indications that in urban areas aerosols make clouds `dirty' and thereby decrease the albedo of the cloud aerosol layer and increase the absorptance of the clouds Kondrat'yev et al., 1981. This effect appears to be quite localized; being restricted to over and immediately downwind of major urban areas, particularly cities emitting large quantities of black soot particles. Kondrat'yev et al.\ noted that the water samples collected from the clouds they sampled were actually dark in color.

A potentially more important impact of aerosol on clouds and climate is that they can serve as a source of cloud condensation nuclei CCN and thereby alter the concentration of cloud droplets. Twomey 1974 first pointed out that increasing pollution results in greater CCN concentrations and greater numbers of cloud droplets, which, in turn, increase the reflectance of clouds. Subsequently, Twomey 1977 showed that this effect was most influential for optically thin clouds; clouds having shallow depths or little column integrated liquid water content. Optically thicker clouds, he argued, are already very bright, and are therefore susceptible to increased absorption by the presence of dirty aerosol. In Twomey's words: ``it an increase in global pollution could, at the same time, make thin clouds brighter and thick clouds darker, the crossover in behavior occurring at a cloud thickness which depends on the ratio of absorption to the cube root of drop nucleus concentration. The sign of the net global effect, warming or cooling, therefore involves both the distribution of cloud thickness and the relative magnitude of the rate of increase of cloud-nucleating particles vis-a-vis particulate absorption.}"

Subsequently, Twomey et al. 1984 presented observational and theoretical evidence indicating that the absorption effect of aerosols is small and the enhanced albedo effect plays a dominate role on global climate. They argued that the enhanced cloud albedo has a magnitude comparable to that of greenhouse warming see Chapter 11 and acts to cool the atmosphere. Kaufman et al.1991 concluded that although coal and oil emit 120 times as many CO2 molecules as SO2 molecules, each SO2 molecule is 50-1100 times as effective in cooling the atmosphere than each CO2 molecule is in warming it. This is by virtue of the SO2 molecules' contribution to CCN production and enhanced cloud albedo.

Twomey suggests that if the CCN concentration in the cleaner parts of the atmosphere, such as the oceanic regions, were raised to continental atmospheric values, about 10% more energy would be reflected to space by relatively thin cloud layers. He also points out that an increase in cloud reflectivity by 10% is of greater consequence than a similar increase in global cloudiness. This is because while an increase in cloudiness reduces the incoming solar radiation, it also reduces the outgoing infrared radiation. Thus both cooling and heating effects occur when global cloudiness increases. In contrast, an increase in cloud reflectance due to enhanced CCN concentration does not appreciably affect infrared radiation but does reflect more incoming solar radiation which results in a net cooling effect.

Moreover, we have seen in Chapter 4 that increases in CCN concentration can reduce drizzle and rain production. Because cloud reflectance is increased both by increased droplet concentrations and by increased column-integrated liquid water, a suppression of drizzle by enhanced CCN concentrations would contribute to enhanced cloud albedo. Albrecht 1989 hypothesized that suppression of drizzle formation would lead to longer-lived clouds which by increasing cloud cover would further enhance the albedo of those clouds. But, this is not necessarily true for all boundary layer clouds. In a marine stratocumulus layer with nearly 100\% cloud cover, the amount of condensate in the form of drizzle is only about 10\% of the total amount of cloud condensate. Thus if drizzle is suppressed, it is not likely to impact cloud cover. Moreover, we have seen in Chapter 4 that drizzle can have complicated feedbacks onto the cloudy boundary layer. In Jiang et al's. 2002 simulations, for example, they found that higher CCN concentrations suppressed drizzle which resulted in weaker penetrating cumulus and an overall reduction in the water content of the clouds. Drizzle, however, destabilizes the boundary layer only when it does not reach the surface. When drizzle settles to the surface, such as in heavier drizzle rate situations or when the drops are larger, the entire boundary layer is cooled and is stabilized Paluch and Lenschow, 1991; Jiang et al., 2002. Thus suppressing drizzle formation can either lead to enhanced boundary layer convection and enhanced cloud albedo, or weaker boundary layer convection and thereby lower albedo clouds.

Moreover, drizzle formation is not controlled totally by concentrations of CCN. Numerous studies Houghton, 1938; Johnson, 1982; Tzivion et al., 1994; Levin et al., 1996; Cooper et al., 1997; Feingold et al., 1999 have indicated that giant CCN GCCN or ultra-giant particles can serve as precipitation embryos and initiate collision and coalescence. These aerosol particles which are greater than five micrometers in radius can form cloud droplets large enough to initiate collision and coalescent regardless of whether they are considered activated drops according to Kohler theory Johnson, 1982. For example, modeling studies of marine stratocumulus clouds by Feingold et al. 1999 showed that increased concentrations of GCCN enhanced precipitation in clouds with moderately high CCN concentrations. However, if CCN concentrations were quite low, the natural precipitation process was so efficient that GCCN had little influence on precipitation.

Thus the susceptibility of the drizzle process in marine stratocumulus clouds to anthropogenic emissions of CCN depends on the presence or absence of large and ultra-giant aerosol particles in the subcloud layer. Over the open sea, the dominant large and ultra-giant aerosol particles are sea-salt particles. While these particles contribute only 10\% or less to the total CCN population, they represent major contributors to the large end of the aerosol size spectrum. Their concentration in the marine boundary layer varies with wind speed, being in greater numbers with stronger winds. { We therefore hypothesize} that the susceptibility of the drizzle process in marine stratocumuli to anthropogenic CCN will depend on wind speed, being less susceptible at higher wind speeds than lower. Another potential source of GCCN is desert dust which has both natural and anthropogenic origins see Chapter 4. But desert dust can also suppress clouds by radiatively heating the cloud layer, and serve as enhanced CCN which

will suppress precipitation. It is not clear how desert dust effects the albedo of marine stratocumulus clouds in a climatological sense because of its potentially complicated impacts.

Let us now examine the evidence supporting or refuting the Twomey hypothesis. The major focus of these studies is on the world's oceans where shallow stratocumulus clouds reside. These shallow clouds, which are believed to be most susceptible to the Twomey effect, cover over 34\% of the world's oceans. Therefore, any consistent trend towards increasing CCN concentration over the oceans has the potential for increased atmospheric albedo and global cooling. Unfortunately, there have not been long-term systematic measurements of CCN concentration anywhere on Earth. We must therefore resort to indirect methods of assessing whether global pollution is affecting climate. In general, oceanic CCN concentrations are low; being on the order of 50 to 100 cm^3.

Over continental areas CCN concentrations range from 500 to 1000 cm3 with some heavily polluted regions reaching several thousand per cubic centimeter. The main source of natural CCN over the oceans is believed to be dimethylsulphide DMS which is excreted by plankton and then liberated into the atmosphere where it is oxidized, probably photochemically, to form non-seasalt-sulphate aerosol Bigg et al., 1984; Charlson et al., 1987; Kreidenweis, 19??

In Chapter 4 we saw that ship tracks Coakley et al., 1987; Scorer, 1987; Porch et al., 1990; Radke et al., 1989 are viewed as a `rosetta stone' which illustrates the interaction of CCN and cloud albedo. The evidence is quite clear that exhaust from some fossil fuel burning ships is producing a plume of aerosol which acts as CCN and thereby enhances droplet concentrations. The enhanced droplet concentrations, in turn, suppress drizzle thus making the clouds wetter. Both the enhanced droplet concentrations and wetter clouds produces a plume or "ship-track" that is brighter than surrounding clouds. Another example is what Rosenfeld 2000 calls "pollution tracks" as viewed by Advanced Very High Resolution AVHRR satellite imagery. These pollution tracks are associated with pollution plumes from specific industrial pollution sources. It is inferred that the "pollution tracks" are clouds composed of numerous small droplets that suppress precipitation. We noted that perhaps the most significant aspect of Rosenfeld's analysis is the conspicous absence of pollution tracks over the United States and western Europe. The implication is that these regions are so heavily polluted that local sources cannot be distinquished from the widespread pollution-induced narrow droplet spectra in those regions.

One way to examine the influence of widespread sources of CCN-generating aerosols on global climate is to use simulations with general circulation models. First it must be recognized that GCMs have grid spacings of 150km to 250km and that clouds often compose a small fraction of the the grid-cell area and the average grid-cell vertical

velocities are very small, approximately 0.01m/s whereas actual cloud-scale vertical velocities are more like 1m/s and often greater. The number of cloud droplets activated in clouds is not only a function of the number of CCN available but also on the peak supersaturations in clouds which is related to cloud-scale vertical velocity. It is, therefore, important to estimate cloud-scale vertical velocities to predict the concentrations of cloud drops. Some modelers have assumed an empirical relationship between predicted sulfate mass concentrations and droplet concentrations Kiehl et al., 2000; Boucher and Lohmann, 1995; Martin et al., 1994, but that is equivalent to assuming there is only a single-value of cloud updraft velocity for all clouds in the model. Others, have estimated vertical velocity from predicted turbulent kinetic energy from their boundary layer models Ghan et al., 1997; Lohmann et al., 1999. This is a step in the right direction, but does not take account of the fact that cloudy updrafts are at the tail of the probability density function PDF of vertical velocity. Chung et al., 1997 assume a normal distribution of vertical velocity with a mean given by the GCM grid point mean.

They then determine the velocity weighted mean droplet concentration which takes into account the tails of their assumed PDF of vertical velocity. But observed PDF's of vertical velocity in the cloudy boundary are found to be multimodal and better fit by double-gaussian PDF's Larson et al., 2001 with a mean that is a function of the RMS vertical velocity not a GCM grid point mean. Moreover, the complications of precipitation or drizzle processes on cloud lifetime, cloud water contents, and cloud radiative properties discussed above cannot be simulated well in GCM cloud parameterization schemes. For example, we have seen that precipitation processes are nonlinear functions of total condensate water contents. As a result GCM model grid-box mean liquid water content is essentially meaningless for representation of precipitation production Stevens et al., 1998b; Pincus and Klein, 2000. As pointed out by Pincus and Klein, a PDF approach to subgrid modeling may be the optimum approach to resolving these deficiencies Larson et al., 2005. Another option, though considerably more computationally expensive is to use what has been called "super-parameterizations" in which cloud resolving models are activated at model grid points Grabowski et al., 2001; Randall et al., 2003\

NOTE: A much more computationally efficient method, which retains the physics of the super-parameterizations, has been proposed Pielke et al., 2005 where look-up tables, or functional fits are used within the parent model while the superparameterization is run off-line.} These models have the capability of predicting cloud-scale vertical velocities and liquid water contents and thus explicitly representing precipitation processes.

Keeping these caveats in mind, let us examine some of the results of GCM simulations of the indirect effects of aerosols on climate. Chuang et al. 1997 used a coupled aerosol chemistry-GCM to examine the influence of anthropogenic aerosols on climate. The model explicitly calculates the conversion of sulfur dioxide gases into sulfate particles. It includes an inventory of natural and anthropogenic emissions of these gases and then predicts the global distribution of the aerosols. Both direct and indirect radiative effects of aerosols are considered. But they do not consider the complexities of drizzle formation and its potential radiative influences. They estimate an indirect radiative forcing ranging

from -0.4 to 1.6 W m^2. They find that the maximum in indirect forcing is over the Atlantic ocean near the coastline of North America. This is in contrast with Boucher and Lohmann 1995 who estimated a stronger magnitude of indirect forcing and especially over polluted land regions. Because aerosol pollution is largely concentrated in the northern hemisphere Schwartz, 1989 surface cooling is concentrated over the North Atlantic and North Pacific oceans due to the higher albedo of contaminated clouds. Several GCM's have thus simulated an alteration in the general circulation which then affects precipitation in areas well beyond those regions Rotstayn et al., 2000; Williams et al., 2001; Rotstayn and Lohmann, 2002. These models were coupled to an ocean mixed-layer model so that enhanced cloud albedo produced cooler ocean surface temperatures in the northern hemisphere. In addition suppressed rainfall resulted in more extensive cloud cover which also cooled ocean surfaces.

The models responded by shifting the Intertropical convergence Zone ITCZ southward which enhanced precipitation in the southern hemisphere tropical regions Rotstayn et al., 2000 and drying in the Sahel zone in Africa Rotstayn and Lohmann, 2002. The latter response is consistent with observed reduction in rainfall in the Sahel zone during the 20th century. Williams et al. 2001 also found a similar response to both direct and indirect effects of pollutant aerosols, and in addition, they found a reduction in the Indian monsoon precipitation during June, July, and August. The cooling in their model also resulted in expanded sea-ice coverage in the Arctic ocean in summer. This is in response to the southward displacement of storm tracks associated with the shift of the ITCZ southward.

Thus the greatest impacts of the enhanced aerosol.concentration were over the north Polar regions and secondarily around 40 degrees north latitude. We would like to note that someone should not interpret the results of those simulations as being quantitative forecasts of the effects of aerosols on patterns and amounts of regional precipitation. As noted previously there are too many uncertainties in the distribution and concentrations of aerosols in the past and even in the present. In addition, we have seen there are many simplifications in the models that limit their ability to realistically simulate indirect effects of aerosols. Instead, these simulations demonstrate the potential that direct and indirect aerosol forcing, even though being regional in nature, can have wide area responses well beyond the regions directly influenced by aerosol changes in radiation.

We have seen that greenhouse warming as a result of enhanced CO2 concentrations is only significant when the global hydrological cycle is enhanced and greater amounts of water vapor are evaporated into the air principally over the oceans but also over land, since water vapor is the dominant greenhouse gas as discussed in Chapter 8. The increased amounts of water vapor in the air, in turn, results in a strong positive feedback to CO2 warming. Recent GCM simulations of both greenhouse warming, and direct and indirect aerosol affects, Liepert etal., 2004 suggest that aerosol indirect and direct cooling reduces surface latent and sensible heat transfer and as a consequence acts to spin-down the hydrological cycle and thereby substantially weaken greenhouse gas warming. This is important since most investigators compare top of the atmosphere radiative differences for greenhouse gas warming and aerosol direct and indirect effects

separately. But since greenhouse warming depends on a spin-up of the hydrological cycle and aerosol direct and indirect cooling counters that, the potential influence of aerosols on climate could be far more significant than previously thought

Aerosols in Mixed-phase Clouds and Climate

We have seen in Chapter 4 that supercooled clouds can exhibit what are called pollution tracks Rosenfeld, 2000 which are believed to be clouds composed of numerous small droplets that suppress precipitation. Perhaps the most significant aspect of Rosenfeld's analysis to global precipitation and climate is the conspicuous absence of pollution tracks over the United States and western Europe. The implication is that these regions are so heavily polluted that local sources cannot be distinquished from the widespread pollution-induced narrow droplet spectra in those regions. We have also seen that pollution can suppress precipitation in wintertime orographic clouds Borys et al., 2000; Borys et al., 2003 by enhancing the concentration of CCN and as a consequence cloud droplets are smaller, which reduces the efficiency of ice crystals collecting supercooled droplets or riming. Further evidence of this effect was suggested in the analysis by Givati and Rosenfeld 2004 of orographic precipitation records downwind of major urban centers in Israel and California. They inferred that precipitation is suppressed by 15 to 25\% downwind of those urban areas.

We have seen that some pollution sources, particularly those associated with mining and heavy metal industries and leaded gasoline are high in ice nuclei IN concentrations Schaefer, 1969. There are indications, however, that there has been a systematic decrease in IN concentrations at several sites in the southern hemisphere as well as Hawaii over the last 25 years Bigg, 1990. It is not known at this time if such observations are a direct result of the increased use of lead-free gasolines or contamination of natural ice nuclei by sulfate-pollutants often called IN poisoning. There are observations, however, of both enhanced CCN and IN concentrations in the boundary layer. For example during the FIRE/SHEBA field experiment in the Arctic basin, Yum and Hudson 2001 found CCN concentrations of 100 and 250 cm^3 active at 1% supersaturation below and above the boundary layer inversion, respectively. At the same time Rogers et al. 2001 measured IN concentrations ranging from approximately 3 L below the inversion to 85 L above the inverion. The air over the ice surface in the Arctic boundary layer is often very clean. But it has long been known that there are major intrusions of polluted air into the Arctic basin and that they often contain high concentrations of cloud condensation CCN Borys and Rahn, 1981; Patterson et al., 1982. Whether the pollution sources are also rich in IN is less well known.

Cloud-resolving simulations of Arctic boundary layer clouds during FIRE-SHEBA, first for a particular day Carri et al., 2005a and then for the entire spring season of the field campaign Carri et al., 2005b were carried out. The model was initialized with either the clean sub-cloud aerosol concentrations throughout the boundary layer or with the observed polluted aerosol concentrations above the inversion and clean below. During the spring season simulations were performed with the model coupled to a sea-ice model. The multi-month simulations were performed using two to three daily SHEBA soundings

nudged into the cloud-resolving model to represent daily variations in the synoptic atmosphere. Mixed-phase clouds prevailed during the first two months of simulation, while predominately liquid clouds were simulated during the last month. The effects of IN entrainment when mixed-phase clouds were present, decreased liquid water paths while ice water paths increased. Even though the above inversion IN concentrations are much lower than estimates from mid-latitudes using the Meyers et al. 1992 formula, the clouds were essentially over-seeded. As a result the crystal fall speeds were reduced and so were the precipitation rates.

This resulted in longer residence times of the ice particles and increased total condensate paths. This produced enhanced downward longwave radiation. Enhanced albedo associated with enhanced CCN concentrations hence dropletconcentrations also occurred but this was much smaller in magnitude so that net surface radiation was increased. As a consequence sea-ice melting rates were greater. Overall the model suggests that the entrainment of a polluted air layer overriding the inversion enhances sea-ice melting rates. Melting rates were approximately 4\% higher when the air above the boundary layer was polluted than when the entire layer is composed of clean sub-cloud air. Only recently have GCMs been able to simulate the effects of enhanced IN concentrations on climate. Lohmann 2002 assumed that a fraction of hydrophilic soot aerosol particles act as contact ice nuclei at temperatures between 0°C and -35°C based on laboratory studies by Gorbunov et al. 2001. She found that increases in aerosol concentration from pre-industrial times to present day pose a new indirect effect, a so-called `glaciation indirect effect', on clouds

She showed that increases in contact ice nuclei in the present-day climate result in more frequent glaciation of clouds and increase the amount of precipitation via the ice phase. This effect can at least partly offset the solar indirect aerosol effect on water clouds and oppose suppression of drizzle by enhanced CCN concentrations. As a result she concluded that precipitation is enhanced by anthropogenic aerosols [Adapted from Lohmann 2002]

It should be noted that whether enhanced IN concentrations increase or decrease precipitation is very cloud specific. It is also the most controversial aspect of cloud seeding. In clouds with low liquid water contents complete glaciation of clouds can overseed them and result in high concentrations of small ice crystals as in Carri et al.'s simulations. On the other hand, modest increases of ice crystal concentrations in some clouds can convert them from a non-precipitating or weakly precipitatiing cloud to one that is precipitating. Unfortunately, neither cloud-scale models or GCMs have demonstrated the ability to predict which regime prevails under a variety of cloud regimes.

Aerosols, Deep Convection and Climate

Most large scale numerical prediction models such as GCMs use convective parameterization schemes such as Arakawa-Schubert 1974, Betts-Miller 1986, or Kuo

1974. These schemes provide heating and moistening by deep convection and have a simple precipitation parameterization that is usually linked to a bulk precipitation efficiency. As such, these models do not have sufficient sophistication in cloud microphysics to include aerosol influences on precipitation or mass fluxes, or heating profiles. An exception, would be those models that use a so-called super-parameterization approach Grabowski et al., 2001; Randall et al., 1984. Since this approach uses cloud-resolving models, it is relatively straight-forward to extend them to include explicit aerosol affects. Using a more classical approach to parameterizing convection, Nober et al. 2003 decreased precipitation efficiency in convective clouds having temperatures warmer than 263 K depending on the cloud droplet number concentration. The instantaneous effects on precipitation formation were very large, reducing precipitation formation by as much as 100\%. However, because these changes are confined to small areas, the large-scale mean precipitation anomalies was not significantly affected.

Khain et al. 2004 postulate that smaller cloud droplets, such as originating from anthropogenic activity, would reduce the production of drizzle drops. When these droplets freeze the associated latent heat release results in more vigorous convection. In a clean cloud, on the other hand, drizzle would deplete the cloud liquid water so that less latent heat is released when the cloud glaciates resulting in less vigorous convection. Thus, they found that a squall line did not form under clean conditions, whereas the squall line developed under continental aerosol conditions which produced more precipitation after two hours. Zhang et al. 2004 came to similar conclusions for different three-week periods over the ARM site in Oklahoma.

In simulations of entrainment of Saharan dust into Florida thunderstorms, van den Heever et al. 2005 also found dust not only impacts the cloud microphysical characteristics but also the dynamical characteristics of convective storms as well. We have seen in Chapter 4 that dust can enhance CCN, GCCN, and IN concentrations. van den Heever et al. considered the influence of dust on deep convection in which dust served as just CCN, then just as GCCN, then just as IN and then the entire combination. In each simulation, dust altered the dynamics of convection by producing greater amounts of supercooled water. But in response to freezing of the greater amounts of supercooled water, the strengthened updrafts thrust more water into anvil levels and produced less accumulated rainfall on the ground by the end of the day. Earlier in the afternoon precipitation was enhanced by dust, but later most of the dust had been washed out by precipitation.

The variations in cloud microstructure and storm dynamics by dust, in turn, alters the accumulated surface precipitation and the radiative properties of anvils. This is in contrast to the `dynamic seeding' concepts in which seeding enhances glaciation of convective clouds which leads to dynamical invigoration of the clouds, larger amounts of processed water, and thereby enhanced rainfall at the ground Simpson et al., 1967; Rosenfeld and Woodley, 1989; 1993. From a radiative perspective it may be more important that the anvil properties of the clouds are modified by dust than surface rainfall. That is, if the direct effects of dust are not so strong that convection is totally inhibited, those storms that do form are likely to produce anvils which are optically thicker and thereby reflect incoming solar radiation and radiate more longwave radiation

to the ground, with the latter response being dominant. Thus dust may act to enhance greenhouse warming.

Overall aerosols have the potential for having substantial impacts on clouds, precipitation, and the radiative properties of clouds. However, the interactions between aerosols and clouds is sufficiently complex that even cloud-resolving models have difficulty in simulating all the complex interactions among physics and dynamics of clouds. Because GCMs cannot resolve all these complexities, it will be some time before we can confidently predict the impacts of aerosols on the global hydrological cycle and on radiation so that we can be confident on the diverse aerosol impacts on global climate.

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