Current Progress in the Study of Global Biogeochemical Cycles

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Abstract
Aspects of global cycles of carbon, nitrogen and sulfur are reviewed. New work defining carbon source and sink strengths in oceans, northern forests and tundra, and wetlands is discussed. Effects of CO$_2$ and N fertilization on the carbon cycle may be large but are currently ill-defined. Recent changes in rates of increase of CO$_2$, CH$_4$ and CO in the atmosphere are probably related to volcanic eruptions. The global nitrogen cycle is grossly disturbed by human activity. Land use change and fertilizer use in the tropics may be major sources of N$_2$O. Ammonia merits further study as a regional pollutant and because of its role in tropospheric aerosol formation. Sulfate aerosols are now recognized as having significant negative forcing on climate. Whilst the direct (radiative scattering) effect of aerosols is well-understood, the indirect effect (altering cloud properties) is very uncertain, and inclusion of aerosol effects in climate models is limited by lack of data on aerosol regional distributions. In the future, industrial growth in developing countries will alter the amounts and global distribution of greenhouse gases and aerosols, and the change and distribution of aerosols will have particularly important implications for future regional climate change.

1. INTRODUCTION

Studies of biogeochemical cycles play an intrinsic part in research programs into climate change. In particular, cycles which involve radiatively active gases and particles have received a great deal of attention in recent years, partly to establish the strengths of sources and sinks and how these strengths are altered by human activity, and partly to investigate the processes by which the cycles are driven. Increasingly, it is being recognized that biogeochemical cycles of elements cannot be treated as entirely independent; improved knowledge of the chemistry which takes place in the atmosphere, soils and the oceans is revealing many ways in which cycles are intricately linked.
In the space available in this paper, we have chosen to concentrate on some of the current issues associated with the biogeochemical cycles of three elements: carbon; nitrogen; and sulfur. Inevitably this assessment is biased by our personal interests, but we have attempted to include examples of some of the exciting recent developments in the atmospheric, oceanic, and terrestrial sciences.

2. THE CARBON CYCLE

Three issues have dominated research into the carbon cycle in recent years. First, balancing the CO₂ budget - establishing the strengths of sources of atmospheric CO₂ arising from human activity and natural systems, and of sinks in the ocean and on the land. Second, understanding feedbacks by which the sources and sinks of a number of carbon trace gases interact with climate change and with increasing CO₂ concentration. Third, most recently, investigating possible causes of sudden changes in the rates of atmospheric accumulation of a number of trace gases that have been observed in the early 1990s.

2.1 Balancing the CO₂ budget

Table 1 shows the global CO₂ budget for the decade of the 1980s as proposed by the IPCC (1994). Emissions are relatively well-established, and occur mainly (90%) in the northern hemisphere from fossil fuel combustion and cement production. Emissions from tropical land use change remain poorly quantified, and improved data from southern Asia, Africa, and tropical South America, preferably collected with a common methodology such as high-resolution satellite imagery, are urgently needed. Until recently it was believed that tundra ecosystems were globally a net accumulator of carbon at a rate of about 0.1 to 0.3 Gt yr⁻¹, but recent work by Oechel et al (1993) suggests that warming in the Arctic may have changed these regions to sources with a global strength of about 0.2 Gt yr⁻¹.

The accumulation of carbon dioxide in the atmosphere is very well defined by a global network of monitoring stations. Analysis of the stable isotope $^{13}$CO₂ shows clearly that the seasonal amplitude in concentration which varies around the globe is dominated by the activity of the terrestrial biosphere in the northern hemisphere, rather than by seasonal changes in fossil fuel emissions or in ocean sink strength. Recent work by Farquhar et al. (1993) on the isotope composition of oxygen in atmospheric CO₂ leads to the possibility of distinguishing influences of different terrestrial biomes in the global CO₂ monitoring network, and when this approach is combined with atmospheric mixing models, it may be possible to resolve some of the present conflict about the relative role of the oceans and the terrestrial biosphere in the net uptake of CO₂.
Table 1
Annual average budget for anthropogenic carbon for 1980-1989
in GtC/yr

<table>
<thead>
<tr>
<th>Sources</th>
<th>GtC/yr</th>
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<tbody>
<tr>
<td>Fossil fuel</td>
<td>5.5 ± 0.5</td>
<td></td>
</tr>
<tr>
<td>Changes in tropical land use</td>
<td>1.6 ± 1.0</td>
<td></td>
</tr>
<tr>
<td>Total emissions</td>
<td>7.1 ± 1.1</td>
<td></td>
</tr>
<tr>
<td>Partitioning to Reservoirs</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Storage in the atmosphere</td>
<td>3.2 ± 0.2</td>
<td></td>
</tr>
<tr>
<td>Ocean uptake</td>
<td>2.0 ± 0.8</td>
<td></td>
</tr>
<tr>
<td>Northern Hemisphere forest regrowth</td>
<td>0.5 ± 0.5</td>
<td></td>
</tr>
<tr>
<td>Other terrestrial sinks (CO₂ and N fertilization, climate effects)</td>
<td>1.4 ± 1.5</td>
<td></td>
</tr>
</tbody>
</table>

Data from IPCC 1994

Estimates of carbon uptake by the oceans have been made by two independent approaches. Methods using radiocarbon, produced naturally in the upper atmosphere or artificially during nuclear weapons testing, as a tracer give larger estimates of uptake by oceans than methods based on air-sea exchange. A recent analysis (Seshahayer et al. 1994) suggests that estimates by the radiocarbon tracer method may need to be reduced by about 25%. The air-sea exchange method, which is based on differences in the partial pressure of CO₂ between the water and the air, and on an exchange function described in terms of wind speed and temperature, has also been reassessed (Robertson and Watson 1992). These authors pointed out that the upper 1 mm or so of the oceans is generally cooler than the bulk mixed layer by about 0.3 °C, and when this thermal skin effect is taken into account, the air-sea exchange method results in CO₂ uptake estimates that must be increased by about 0.7 Gt C yr⁻¹. Thus recent work has brought the two approaches into much closer agreement in defining ocean uptake.

There has been debate about whether increases in ocean phytoplankton productivity may have increased the ocean sink strength for CO₂ over the last 100 years or more. Analysis by Falkowski and Wilson (1992) of changes in phyto-plankton biomass in the north Pacific Ocean over the last 70 years shows that changes are too small to have a significant effect on the sink strengths for atmospheric CO₂. Although there are very few historical data for other main ocean basins, it seems likely that this conclusion applies on a global scale. In contrast to the open ocean, coastal zones have undoubtedly experienced large increases in nutrients associated with human population changes, but Falkowski and Wilson concluded that there is no conclusive evidence yet to suggest that these coastal zones represent a significant new sink.
As an example of interacting biogeochemical cycles between the atmosphere and ocean, it has been proposed that, because iron concentrations limit phytoplankton productivity in some parts of the ocean, deposition of iron from either human activities or volcanic eruptions might increase the ocean sink strengths for CO₂. An experiment has recently been conducted in the equatorial Pacific Ocean to test this hypothesis, and Watson et al. (1994) reported some early results. When a small patch (8 x 8 km) was enriched with iron, there was a significant depression in the surface concentration of CO₂ within 48 hours of the iron release, but the effect was only a small fraction (about 10%) of the CO₂ drawdown that would have occurred if the enrichment had resulted in the complete utilization of all other available nutrients. Reasons why the fertilization procedure was much less effective in the open ocean than in the laboratory are not yet clear, but at present the results do not support the idea that iron fertilization significantly affects the oceanic CO₂ sink strength.

The weight of evidence at present suggests that in order to balance the sources of CO₂ in Table 1 against sinks and atmospheric storage, there must be an ill-defined terrestrial sink in northern temperate latitudes. This conclusion is reached by inversion of the observed atmospheric CO₂ distribution combined with atmospheric tracer models, making constraining assumptions about ocean and terrestrial sinks (Tans et al. 1990, Enting and Mansbridge 1991). The likeliest terrestrial processes contributing to this sink are: changing forest management; and enhancement of productivity due to atmospheric CO₂ increases and/or nitrogen fertilization from atmospheric deposition.

A number of recent carbon inventories of temperate forest systems have concluded that such systems may have been a sink for about 0.5 GtC/yr in the last 20 years or so, partly because of natural regrowth and replanting after forest harvesting, and partly through fire suppression (IPCC 1994, Auclair, personal communication). Although there is much evidence from laboratory and controlled environments to show that plant productivity can be increased by 30% to 40% when CO₂ is doubled, there is no conclusive evidence from the field to show long-lasting increases in northern temperate ecosystem productivity in response to increased CO₂. There are sound biochemical reasons indicating that the interaction of CO₂ and temperature is such that the benefits of CO₂ fertilization cannot be achieved at low temperatures (Long, 1991). This may explain partly why, when Oechel et al. (1994) exposed natural arctic tundra to doubled CO₂ concentration, there was no long-term boost in carbon sequestration. An alternative explanation is that, after an initial burst of productivity, plants exhaust the supply of soil nutrients, and this limits future productivity. There have been insufficient long-term experiments with perennial systems such as forests and tundra to determine whether the equilibrium long-term response involves eventual changes in soil nutrient availability that would allow productivity to be enhanced. IPCC (1994) estimated that CO₂ fertilization may have accounted for a sink of 0.5 to 2.0 GtC/yr during the 1980s, but such estimates must be regarded as extremely tentative.
In and around industrialised regions, ecosystems may receive substantial inputs of nitrogen, arising from fossil fuel burning and agriculture, and this input can act as a fertilizer. IPCC (1994) speculated that this fertilizer effect could have increased terrestrial carbon storage by 0.2-1.0 GtC/yr in the 1980s. One of the most important current programs aimed at understanding the interaction between boreal forests and the atmosphere is the international multi-agency BOREAS Program taking place in northern Canada (Sellers, et al. 1995). Analysis of the many detailed records of CO₂ exchange collected during BOREAS should help in quantifying the scale of net carbon input to northern forest systems.

2.2 Feedback processes

We have already mentioned a number of studies which indicate interactions between changing climate and atmospheric CO₂ concentrations and the net exchange of CO₂ between the surface and the atmosphere. There has also been interest in the sensitivity of methane fluxes to climate change and CO₂ concentration. Whiting and Chanton (1993) found that, for wetlands of varying productivity around the world, higher net primary production was associated with higher emissions of methane. It has therefore been suggested that, if CO₂ increased the productivity of wetland vegetation, some of the benefits of carbon sequestration would be lost because of increased methane emissions. Dacey et al. (1994) recently presented results supporting this view. They studied methane emissions from a marsh that had been exposed in open-top chambers to twice ambient atmospheric CO₂ for the previous 7 years, and found that methane emission over a 10-day period from the CO₂-enriched sites was nearly 80% higher than in control sites. If confirmed in longer-term work, the implications of this observation are important, not only for methane fluxes from natural ecosystems but also for fluxes from wetland rice production where much effort is put into increasing productivity.

Most soils that are not flooded consume methane, but the extent varies with soil water content and land use. A number of recent reports have shown that inputs of nitrogen in the form of ammonium to soils strongly inhibit soil methane consumption (King and Schnell 1994). Ammonium concentrations in many soils have increased in recent years as a result of land use changes and increases in the ammonium concentration of precipitation. Similar responses are not observed in soils treated with nitrate-based fertilizer or farmyard manure (Goulding et al., in press). Goulding et al also analyzed soils from long-term experiments at Rothamsted, England, and showed that extended (150 years) cultivation of land for arable crops reduced methane uptake rates by 85% compared to those in soil under calcareous woodland. King and Schnell argued that past increases in atmospheric methane concentration may have increased the inhibitory effect of ammonium on soil methane uptake, and this mechanism would provide a positive feedback on future atmospheric methane concentrations.
2.3 Recent changes in atmospheric accumulation of trace gases

One of the most puzzling and yet instructive aspects in the study of trace gas biogeochemistry occurred in the early 1990s. Until this time, CO₂ concentrations around the world had increased rather consistently over the previous 30 years at about 0.5 - 1.5 vpm per year, with the rate tending to increase with time (Fig. 1).

After mid-1991, continuing throughout 1992 and 1993, the growth rate of atmospheric carbon dioxide slowed by an unprecedented amount (Keeling, 1993, Sarmiento, 1993). Concentrations of carbon monoxide CO and methane CH₄, which had also been increasing steadily up to 1991, grew slowly from 1991 to 1993 (Fig 2). The cause of these large changes was almost certainly the eruption of Mount Pinatubo in the Philippines in June, 1991, but the mechanisms that brought about the changes are a matter for debate. As we discuss later, it seems likely that the carbon dioxide anomaly is associated with the volcanic aerosols that were injected into the stratosphere, reducing solar radiation at the surface and producing cooling on a global scale. Cooling, and
perhaps associated changes in rainfall and evaporation, could alter the balance between photosynthesis and respiration on the land, and the sink strengths of the ocean for carbon dioxide.

![Graph showing CH4 concentration](image)

**Figure 2** Globally averaged CH$_4$ concentration showing low growth rates during 1992 and 1993. (From IPCC 1994)

The changes since 1991 in atmospheric methane and carbon monoxide are probably even more complex, because both gases have a major atmospheric sink by reaction with the hydroxyl radical OH. A recent analysis by Bekki et al. (1994) suggested that an unprecedentedly large depletion of stratospheric ozone over this period may have contributed to the sharp decrease in growth rates of both gases. The decrease in stratospheric ozone would allow more ultraviolet radiation to reach the troposphere and this would have resulted in increased concentrations of OH. Bekki et al. concluded that they could account for almost half of the 1992 decrease in growth rates of both gases by this mechanism, but there may also have been changes in source and sink strengths at the surface to account for the remainder. It seems likely that the ozone depletion in the stratosphere was also caused by the Pinatubo eruption, because the fine aerosol of sulfuric acid droplets resulting from the injection of 15 to 20 million tonnes of sulfur dioxide into the stratosphere interacted with other stratospheric chemicals to destroy ozone.

The consequences of this natural event provide an excellent test of our understanding of the carbon cycle, and serve as a reminder of the complex interactions that are contained within the carbon cycle.
3. THE NITROGEN CYCLE

In comparison to the global carbon cycle, the global nitrogen cycle has been much more grossly disturbed by human activities. Table 2 summarizes annual terrestrial fluxes prior to substantial human alteration, and lists current changes arising from human activities.

Table 2
Global fluxes of nitrogen (in Tg/yr) in unperturbed terrestrial ecosystems and as a consequence of human activity

<table>
<thead>
<tr>
<th></th>
<th>TgN/yr</th>
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<tbody>
<tr>
<td><strong>(i) Unperturbed systems</strong></td>
<td></td>
</tr>
<tr>
<td>Biological N fixation</td>
<td>100</td>
</tr>
<tr>
<td>Fixation by lightning</td>
<td>10</td>
</tr>
<tr>
<td>Denitrification</td>
<td>90</td>
</tr>
<tr>
<td><strong>(ii) Perturbed systems</strong></td>
<td></td>
</tr>
<tr>
<td>Biomass burning</td>
<td>40</td>
</tr>
<tr>
<td>Tropical land clearance</td>
<td>22</td>
</tr>
<tr>
<td><strong>(iii) New fixation</strong></td>
<td></td>
</tr>
<tr>
<td>Manufacture of fertilizer N</td>
<td>80</td>
</tr>
<tr>
<td>N fixation by legume crops</td>
<td>30</td>
</tr>
<tr>
<td>N fixation by fossil fuel combustion</td>
<td>25</td>
</tr>
</tbody>
</table>

Data from Vitousek and Matson, 1995

Since the late 1970s, the production of nitrogen fertilizer has probably increased by about one-third (Vitousek and Matson, 1995). Prior to that time, most of this fertilizer was used in developed countries of the temperate zones, but since then, the rate of increase in nitrogen fertilization has been extremely rapid in the tropics, so that by the late 1980s, more than half of the global nitrogen fertilizer use was in the developing world (including China). This trend is likely to continue, with the implication that the distribution of trace gas fluxes to the atmosphere, discussed in the following sections, will change substantially.

Changes in the source strengths of two trace gases as a result of the disturbed nitrogen cycle have attracted particular attention in recent years. Nitrous oxide emissions are influenced by fertilizer use and land use changes; and ammonia arising from both fertilizer use and animal production has been shown to create important regional problems, especially in Europe.
3.1 Nitrous Oxide

The global budget of nitrous oxide has been revised recently by the IPCC (1992), based on new information on soil fluxes from tropical ecosystems and temperate forests, further work on cultivated soils, and new estimates of emissions from biomass burning. Large tropical sources are required to explain the N$_2$O latitudinal gradient revealed by atmospheric monitoring over the last ten years. Keller and Matson (1994) recently reviewed sources of N$_2$O in the tropics and evaluated the effects of land use changes. Wet undisturbed tropical forests appear to account for the largest natural terrestrial source, and this is associated with the large rates of nitrogen transformation in the soil and cycling through vegetation in these systems. Even so, it seems necessary to include additional tropical sources to balance the global budget, and Keller and Matson proposed that tropical land use change and intensification of tropical agriculture may be significant contributors towards this missing source. In particular, the creation of young pastures from previously undisturbed systems seems likely to considerably increase N$_2$O fluxes to the atmosphere, and the increasing use of nitrogen fertilizer in tropical systems appears from the few measurements available to cause larger fluxes of N$_2$O than would be found from similar practice in temperate crop systems. It seems likely that crop and soil management practices can be manipulated to control nitrous oxide flux and there is clearly a need in temperate and tropical areas for further work to explore this possibility. Since nitrogen fertilization increases emission of nitrous oxide to the atmosphere and may decrease absorption of methane by soils (see earlier), the potential of improved soil management for slowing the buildup of radiatively active trace gases in the atmosphere is very important.

3.2 Sources and Sinks of Ammonia

Concern about ammonia fluxes from intensive agriculture in Europe and eastern North America arises principally because dry and wet deposition of reduced nitrogen compounds can make a substantial contribution to the acidification and nitrogen eutrophication of semi-natural systems (Fowler et al. 1989). The importance of ammonia fluxes in global radiative forcing has not been adequately explored yet, but reactions between ammonia and sulfur dioxide to create ammonium sulfate aerosols, discussed in the next section, are an important contributor to the global anthropogenic aerosol burden.

One of the most important advances in recent years has been the development of micrometeorological methods for studying ammonia fluxes between vegetation and the atmosphere. These techniques have allowed investigations over grazed pastures, fertilized agricultural crops, and natural ecosystems. Sutton et al. (1995) recently reviewed this work. The measurements have clearly demonstrated that there is an NH$_3$ 'compensation point' associated with plant tissue, so that emission from plants occurs when atmospheric concentrations are below the compensation point, and deposition occurs at
higher atmospheric concentrations. Figure 3 summarizes measurements over agricultural crops and semi-natural vegetation (Sutton et al. 1995), showing that at low NH₃ concentrations, agricultural crops are a source of NH₃ to the atmosphere and semi-natural vegetation is a sink. The figure also indicates that the compensation point is different for the two systems, as would be expected as a result of nitrogen fertilization of crops. It seems likely that natural vegetation on which there has been substantial NH₃ deposition could also have a higher compensation point, and so the sink strengths of natural vegetation for NH₃ may decrease in polluted regions with time.

![Figure 3](image)

Figure 3 Micrometeorological measurements of the variation of NH₃ fluxes (positive = away from the surface) with NH₃ concentration over agricultural crops and semi-natural ecosystems (From Sutton et al., in the press)

The global ammonia budget has been relatively neglected in studies of the nitrogen cycle. Schlesinger and Hartley (1992) concluded that the major uncertainties and emissions were associated with fluxes from undisturbed soils. They also particularly commented that deposition of ammonium in rain in the eastern United States declined over the 20-year period from 1963 to 1982, which they regarded as surprising, given the increasing use of urea fertilizer. They concluded that atmospheric interactions with sulfur dioxide to form sulfate
aerosol may account for this observation, but it also seems likely from recent work that ammonia deposition on natural vegetation close to agricultural fields has substantially increased, and the consequences of this nitrogen input deserve more attention.

4. THE SULFUR CYCLE

The global sulfur cycle has been severely perturbed by human industrial activities for many years. Because world industrial activity and fossil-fuel consumption have been concentrated in the northern hemisphere, there are virtually two global sulfur cycles: a relatively "natural" cycle in the southern hemisphere, and a vastly perturbed cycle in the northern hemisphere. Some consequences of the perturbed cycle, such as acid rain, have been recognized for decades. But we have only in the past few years begun to appreciate the potential importance for global climate of human impacts on the sulfur cycle. In the following sections we will briefly review recent findings about the climatic roles of both the natural and perturbed sulfur cycles.

4.1 Sulfur, aerosols, and climate

Scattering of solar radiation by atmospheric aerosols effects the atmospheric radiation balance because it reduces the amount of energy from the sun that reaches the surface of the earth. Scattered radiation may be either lost to space or absorbed by aerosols, but both processes result in a net loss of energy to earth's surface and therefore net cooling. Thus, aerosol scattering has a negative forcing influence on climate - the opposite of greenhouse gases such as CO$_2$.

Although the mass of sulfur in the atmosphere is only about $10^{-5}$ that of carbon, it dominates scattering in the atmosphere. This is because sulfur oxidizes and hydrates to form sulfate aerosol particles that are in the size range (0.1 - 1$\mu$m) that scatters visible radiation very effectively. Such scattering is roughly $10^5$ times more efficient per atom as a climate forcing mechanism than the radiation absorption of greenhouse gases such as CO$_2$ (Shaw, 1983), but the radiative influence of aerosols on climate is reduced by their much shorter atmospheric residence time, less than a week, compared to an effective residence time of decades for CO$_2$.

Sulfate aerosols contribute to radiation scattering in two distinct ways: by scattering directly and by modifying cloud optical properties and thereby influencing radiation scattering by clouds. The latter "indirect" effect has been appreciated as an important feature in global climate for some years, but is an extremely complex process and is still poorly understood.
4.1.1 The natural sulfur cycle and climate

We have surprisingly little quantitative understanding of the role of the natural sulfur cycle in climate. This is partly because the subject has only recently attracted much attention, and partly because the natural cycle has been so perturbed by human activity. There are few data on the distribution of sulfate aerosols even today, much less from prior eras, and trying to find the climate signal of natural sulfate aerosols in a vastly perturbed world is extremely difficult. Nonetheless, there are several likely mechanisms by which the natural sulfur cycle plays a role in climate.

The marine dimethyl sulfide (DMS) effect - the brightening of marine stratus clouds hypothesized by Charlson et al. (1987) - may have been the dominant process by which the natural sulfur cycle affected climate before the industrial revolution. An intriguing aspect of this hypothesis is the suggestion of a potential feedback loop which, if negative, would act to stabilize climate. However, studies of ice-core concentrations of methane sulfonic acid and non-seasalt sulfate (atmospheric oxidation products of DMS) (Legrand et al., 1988) suggest instead that there was a positive feedback during previous ice ages.

To date, evidence supporting the marine DMS hypothesis has been difficult to gather (Ayers and Gras 1991, Ayers et al. 1991, Bates et al. 1987) because of the difficulties associated with measuring simultaneously all the necessary variables (Bates et al., 1990) and the interference from anthropogenic sulfate aerosols (Falkowski et al. 1992, Schwartz 1988). Field studies planned for 1995 - 96 in the southern Pacific ocean are aimed at determining whether the 'cloud brightening' mechanism actually exists, and its potential strength.

Volcanic eruptions (e.g. Mt. Pinatubo in June, 1991 in the Philippines) have important impacts on the natural sulfur cycle by injecting huge pulses of sulfur into the stratosphere which oxidize to form sulfate aerosols. Unlike tropospheric aerosols, these stratospheric aerosols remain in the atmosphere for several years due to the lack of aqueous removal mechanisms in the extremely dry stratosphere. They therefore achieve circumpolar distributions and may cause global cooling for a significant period. Such aerosols also deplete stratospheric ozone, as discussed earlier.

4.1.2 Anthropogenic changes to the S cycle

Sulfur is an integral element in all biological materials, and all biogenic oil and coal contain approximately 1% - 10% S by mass. Therefore, production of sulfur gases is an inevitable by-product of fossil fuel combustion.
Emissions of sulfur to the atmosphere from human activities are now 2 - 3 times natural emissions annually. In the past few decades, the major areas of sulfur flux to the atmosphere have been the eastern United States and western and central Europe, and globally, about 94% of the emissions are in the northern hemisphere (Schwartz 1988). Because aerosols are removed from the atmosphere before they can be transported across the equator, this leads to vastly different distributions of aerosols in the northern and southern hemispheres (Langner et al. 1992). The sulfate haze that blankets much of the northern hemisphere is now recognized to be a direct result of fossil fuel burning rather than from natural sources. Further evidence for the disparate hemispheric cycles comes from 200% - 300% increased sulfate deposition to Arctic ice but not to Antarctic ice in the past hundred years (Mayewski et al. 1990). Because no global or even regional aerosol sampling network exists, and satellite observations are lacking, our best estimates of aerosol distributions come from computer models which begin with known sources and simulate atmospheric transport, chemistry, and physics to predict aerosol distributions (Langner et al. 1992).

Although the mechanism of cooling by direct aerosol scattering is relatively simple, its impact was until recently underestimated, largely because it was not realized how much of the sulfate aerosol haze in the northern hemisphere is actually from industrial emissions. Although one recent estimate suggests that perhaps less than 10% of sulfur emissions result in the formation of new aerosol particles (Langner et al. 1992), the rate of new sulfate particle formation may have doubled since pre-industrial times.

Recent re-evaluations of the direct climate forcing by radiation scattering from anthropogenic aerosols have suggested cooling of similar magnitude to the CO₂ warming (Charlson et al. 1992, IPCC 1994), leading to speculation that the "greenhouse signal" predicted in the late 1980's has been partially masked by concomitant sulfate aerosol production. Because the cooling due to aerosol scattering is localized, it is thought to be heavily concentrated around eastern North American and central Europe.

Aerosol modification of cloud albedo (indirect climate forcing by aerosol) is a much more difficult problem. The distribution and radiative properties of clouds are probably the major uncertainties in climate prediction models, and there are no models today which treat clouds in a wholly realistic manner. Worse, the increased reflectivity of clouds from sulfur aerosol condensation nuclei is extremely non-linear and poorly understood. Nonetheless, several recent attempts to estimate the impact of human sulfur emissions on cloud properties (Jones et al. 1994, Wigley 1989) suggest cooling which may be similar to that produced from aerosol scattering - that is, roughly comparable to CO₂ warming.
Climate models are only just beginning to include sulfur emissions and direct and indirect effects of aerosols (Jones et al. 1994, Kiehl and Briegleb 1993, Taylor and Penner, 1994, Wigley 1989). Initial results suggest that predicted climate responses when aerosols are included may be quite different than for radiative gases alone. Feedbacks within the climate system may lead to cooling not just in regions of sulfur emission but also in far-removed areas, such as in the sub-Arctic (Taylor and Penner, 1994). One consequence of the northern-hemisphere enhancement of sulfur aerosols is that warming associated with increases in greenhouse gases may occur more quickly in the southern hemisphere, where it is not partially offset by aerosol cooling.

5. CONCLUDING REMARKS

Changes in global economic and social systems are likely to have profound effects on emissions of radiatively active gases and particles in future decades, with implications for global climate change. The breakup of the Soviet Union, explosive growth in the third world, and the economic emergence of Asia, in particular the "industrial revolution" in China, will lead to geographically changing patterns of fuel consumption over the next decade that are unlike anything in the past 25 years. Carbon and sulfur emissions from rapidly industrializing nations are likely to soar. At the same time, emissions in the developed world may decrease, as more stringent controls take effect. In general, we may expect an increase in emissions from low latitudes on both sides of the equator, and a possible stabilization in the higher-latitude emissions from North America and Western Europe. Emission changes in Eastern Europe and the countries of the former Soviet Union are major uncertainties.

Geographic patterns of fossil fuel emissions of greenhouse gases are not particularly important, since these species are well mixed around the globe and have lifetimes of years to decades in the atmosphere. Although climate response to greenhouse gas forcing will certainly vary between regions, the distribution of responses is likely to be relatively insensitive to where the gases are emitted.

The situation is very different for short-lived sulfur aerosols. Their climatic effects are intrinsically regional, since they do not exist in the atmosphere long enough to be dispersed globally. Consequently, changes in the distribution of sulfur emissions will result in different local climatic impacts. However, it is likely that regional climate response will not be limited only to areas of strong forcing, because of feedbacks in the climate system (Taylor and Penner 1994).

We have barely begun to explore the complex interactions between the climatic forcing of industrial carbon, sulfur, and other emissions, as well as our other diverse impacts on global biogeochemical cycles. If world economic changes are more rapid than scientific advances necessary to understand the climatic effects of these coupled emissions, we may be chasing a "moving target" of
climate forcing. Climatic change policy decisions based on today’s economic and social scenarios may be wrong for tomorrow’s world unless we understand the effects on climate of our modifications of the major biogeochemical cycles.

6. REFERENCES


