Atmospheric Deposition; Atmospheric Models

OCN 401 - Biogeochemical Systems

28 August 2012

Reading: Schlesinger, Chapter 3

Outline

1. Atmosphere Fundamentals
    - Structure
    - Mixing
    - Aerosols

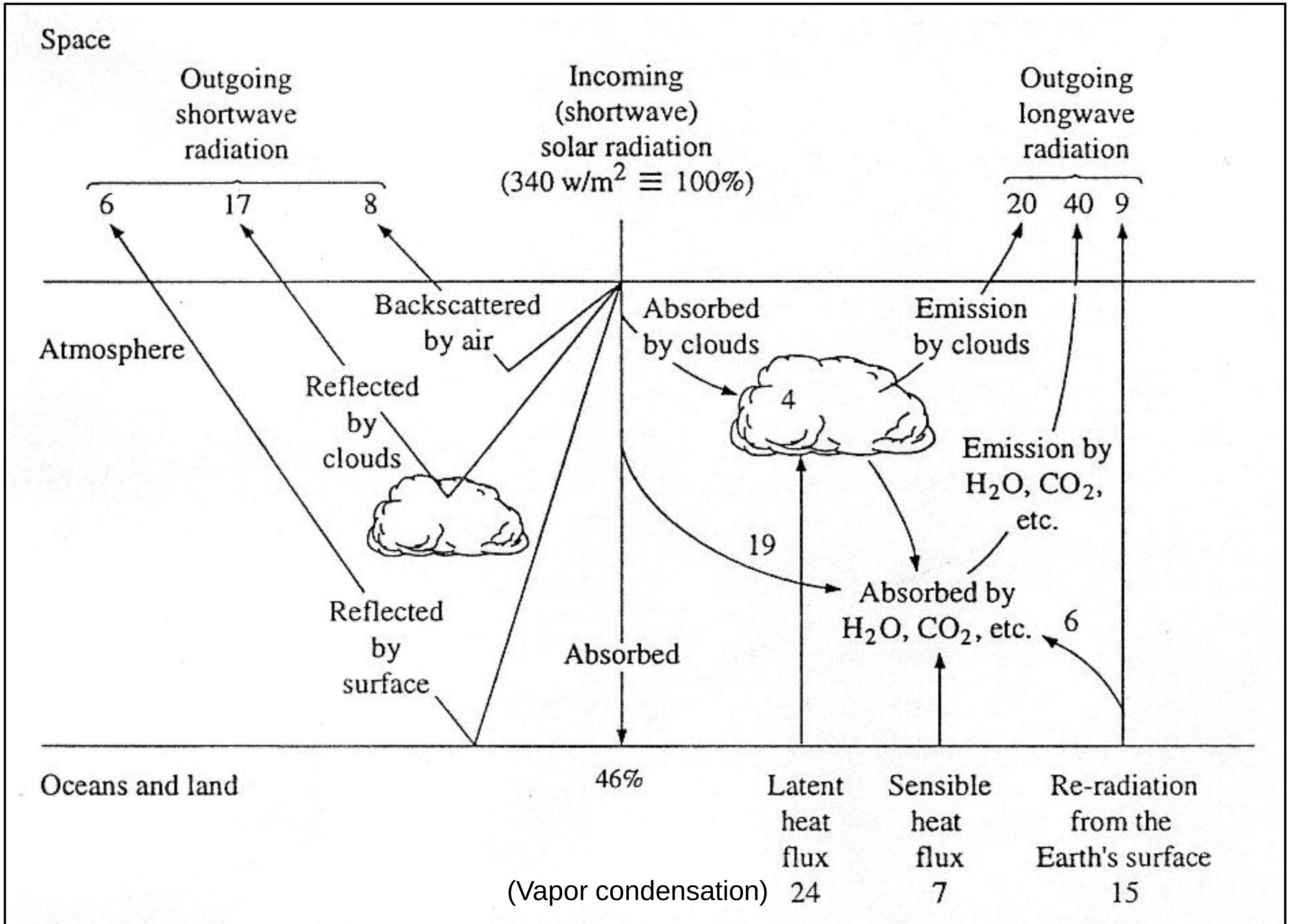
2. Atmospheric Deposition
    - Wet deposition
    - Dry deposition
    - Gas absorption
    - Regional patterns of deposition

3. Atmospheric Models
    - Model classification
    - Limiting factors on model accuracy
    - Consensus results
    - Cooling effects due to aerosols

4. Use and Abuse of Climate Models

Thermal Structure of Atmosphere

~50% of Sun's energy penetrates the atmosphere and is absorbed at surface of the Earth and re-radiated



Atmospheric Mixing

- Tropospheric mixing time ≈ few months
- Inter-hemispheric mixing of tropospheric air ≈ 1 yr
- Thus, inter-hemispheric gradients imply large sources

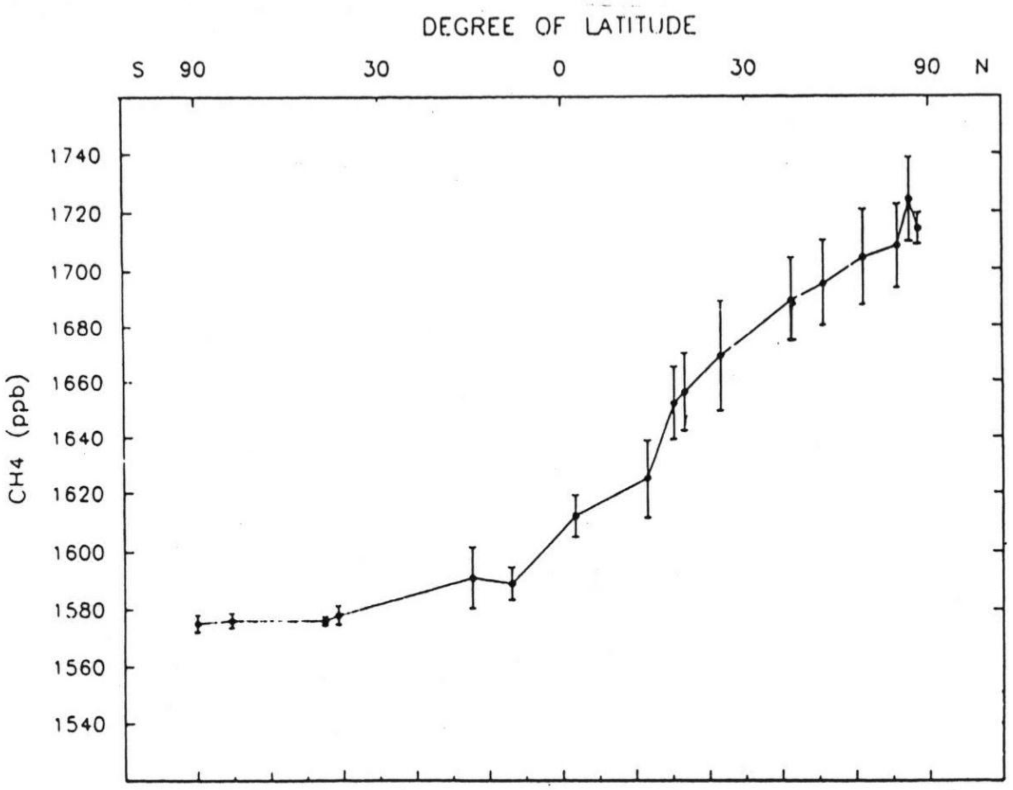
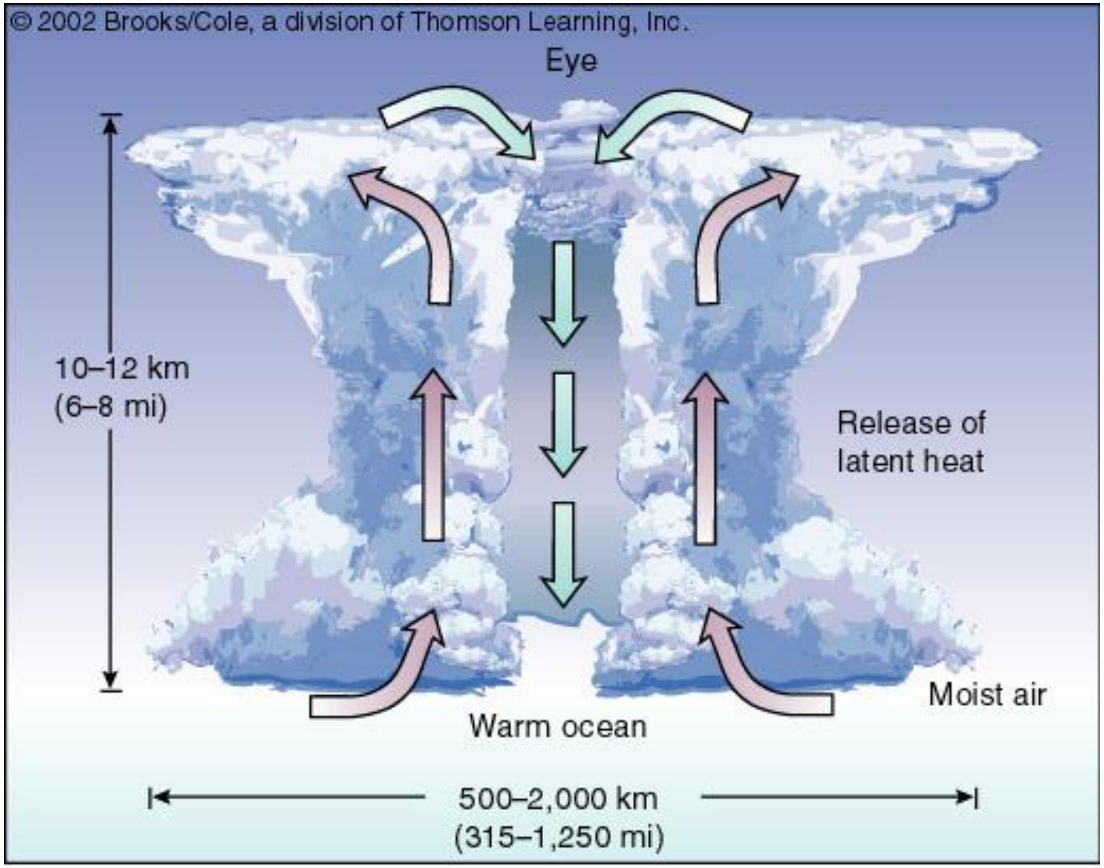


Figure 3.4 The latitudinal variation in the mean concentration of CH_4 (methane) in Earth's atmosphere. From Steele et al. (1987).

Transfer Between Troposphere and Stratosphere

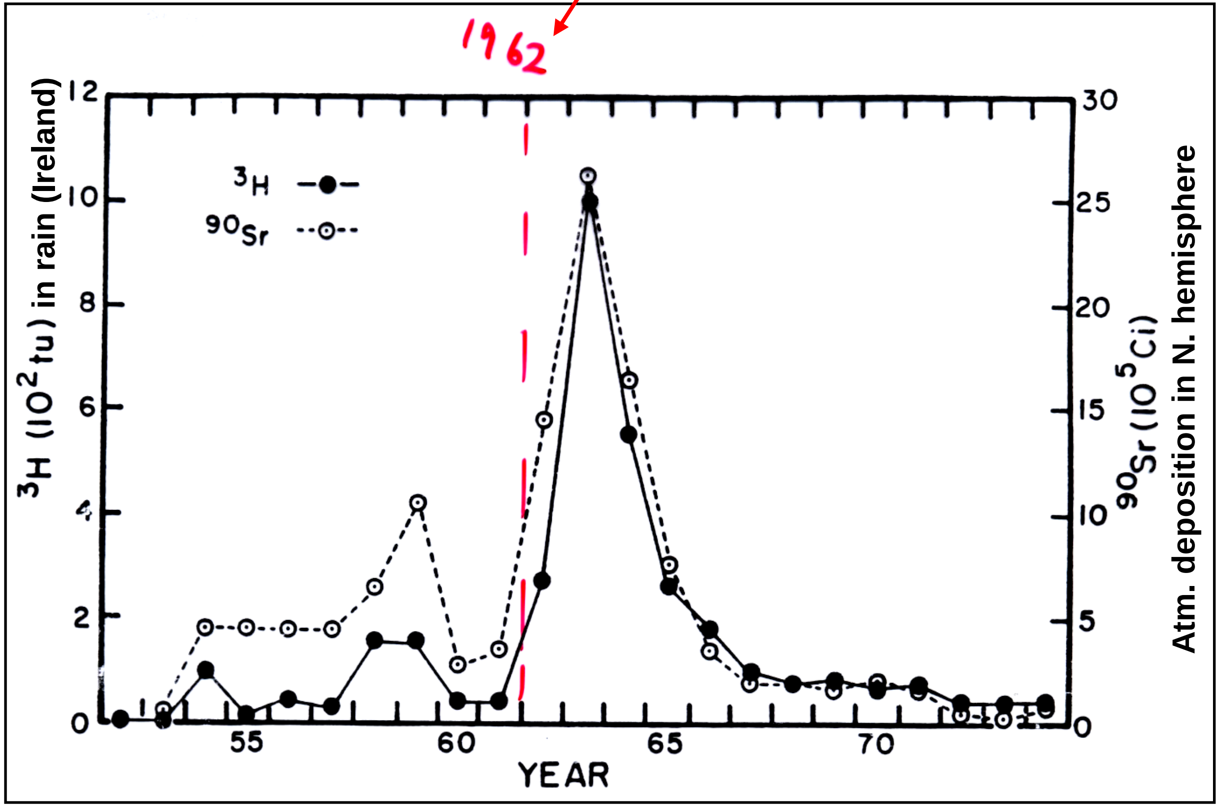
Dominated by rising air masses in tropics

Extreme case: hurricanes
Carry tropospheric air into stratosphere



Only limited mixing between stratosphere and troposphere, at tropopause -- known from mixing of products of atmospheric weapons testing:

End of atm testing (*i.e.*, input to stratosphere)



Calculate this mixing using residence-time calculations

Aerosols Types and Sources

Aerosol: very small particle of solid or liquid suspended in a gas

Range of size:

- From <10 nm to >100 μm in diameter
- From a gathering of a few molecules to the size where the particles no longer can be carried by the gas

Some of these particles are emitted directly to the atmosphere (*primary emissions*)

Some are emitted as gases and form particles in the atmosphere (*secondary emissions*)

Soils (dust)

Soil blown into atmosphere → large transport of continental material

But how much?

1500 x 10^{12} g/yr (Schlesinger)
910 x 10^{12} g/yr (Duce)
360 x 10^{12} g/yr (Prospero)

Differ by a factor of 4+!

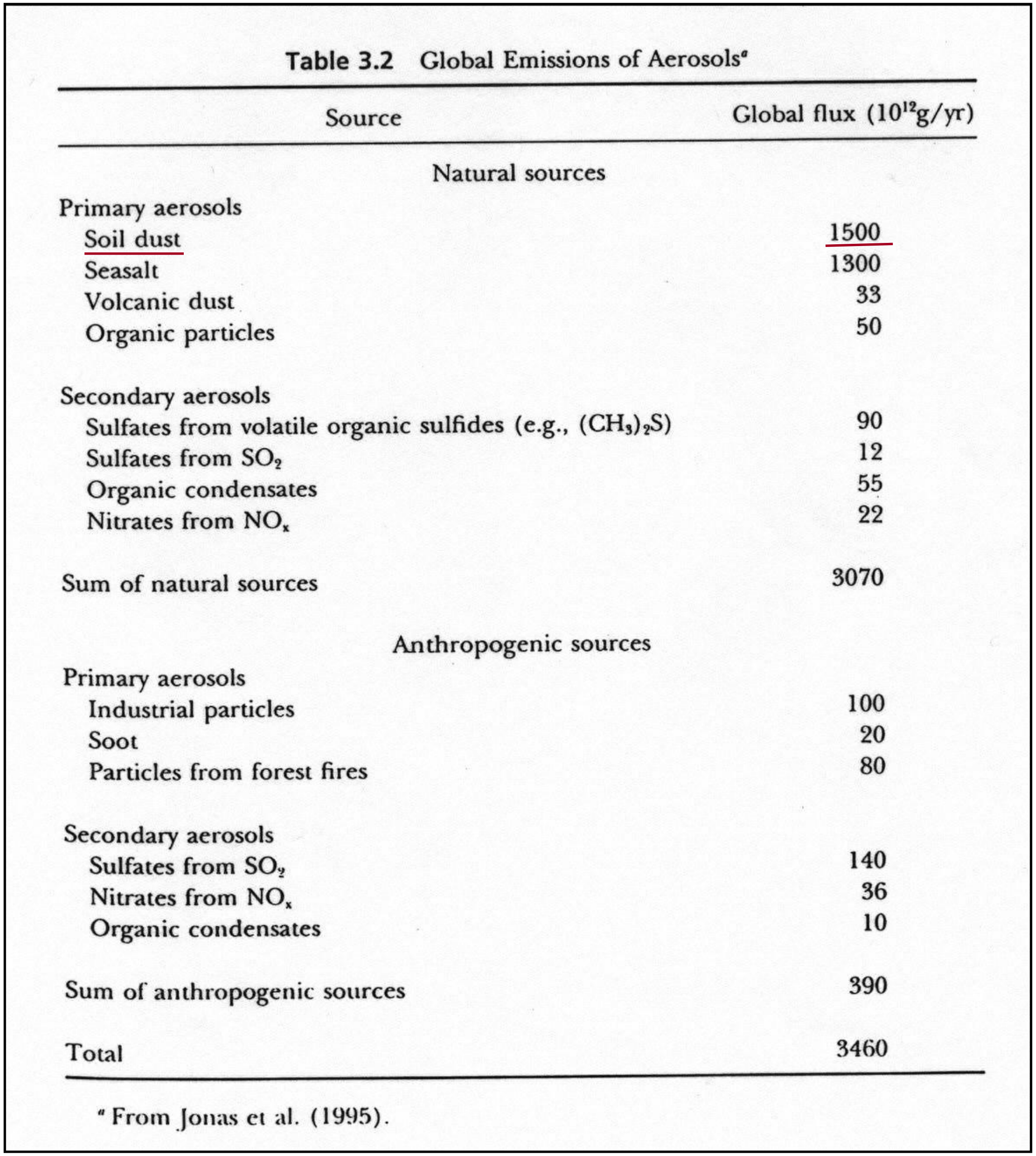


Table 3.2 Global Emissions of Aerosols[a]

Source	Global flux (10^{12}g/yr)
Natural sources	
Primary aerosols	
Soil dust	1500
Seasalt	1300
Volcanic dust	33
Organic particles	50
Secondary aerosols	
Sulfates from volatile organic sulfides (e.g., $(CH_3)_2S$)	90
Sulfates from SO_2	12
Organic condensates	55
Nitrates from NO_x	22
Sum of natural sources	3070
Anthropogenic sources	
Primary aerosols	
Industrial particles	100
Soot	20
Particles from forest fires	80
Secondary aerosols	
Sulfates from SO_2	140
Nitrates from NO_x	36
Organic condensates	10
Sum of anthropogenic sources	390
Total	3460

[a] From Jonas et al. (1995).

Sea spray

Water droplets evaporate and produce large particles that settle quickly

Fluxes not well known (~1-10 x 10^{15} g/yr)

Returns Cl to land

Volcanic Sources

Source of soils downwind of eruptions

Violent eruptions put material in stratosphere. Global dispersion, long residence time.

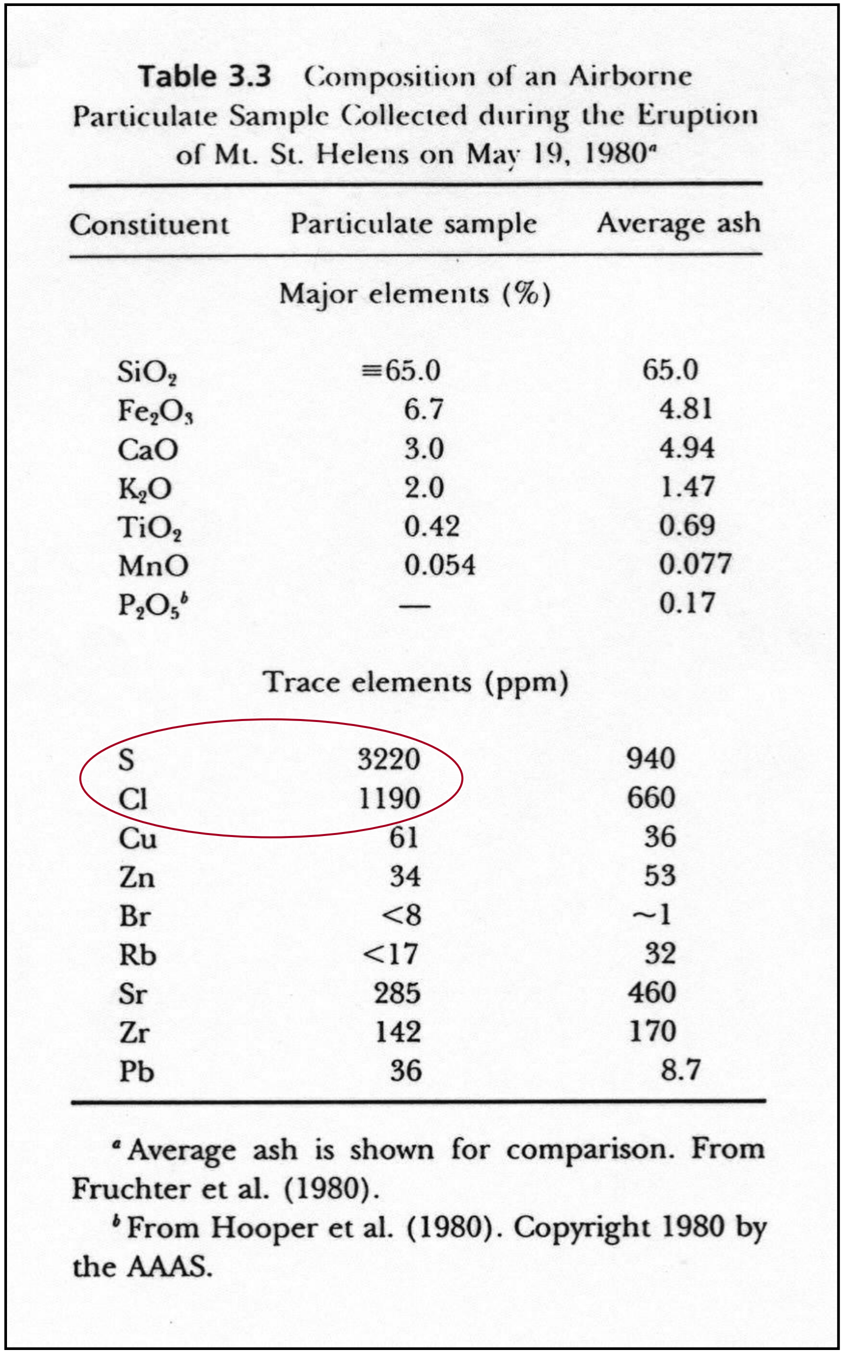


Table 3.3 Composition of an Airborne Particulate Sample Collected during the Eruption of Mt. St. Helens on May 19, 1980[a]

Constituent	Particulate sample	Average ash
	Major elements (%)	
SiO_2	≡65.0	65.0
Fe_2O_3	6.7	4.81
CaO	3.0	4.94
K_2O	2.0	1.47
TiO_2	0.42	0.69
MnO	0.054	0.077
P_2O_5[b]	—	0.17
	Trace elements (ppm)	
S	3220	940
Cl	1190	660
Cu	61	36
Zn	34	53
Br	<8	~1
Rb	<17	32
Sr	285	460
Zr	142	170
Pb	36	8.7

[a] Average ash is shown for comparison. From Fruchter et al. (1980).

[b] From Hooper et al. (1980). Copyright 1980 by the AAAS.

Soot

- Mostly from forest fires and fossil fuel burning

- Biomass burning ≈ 1 x10^{13} g/yr from Amazon alone

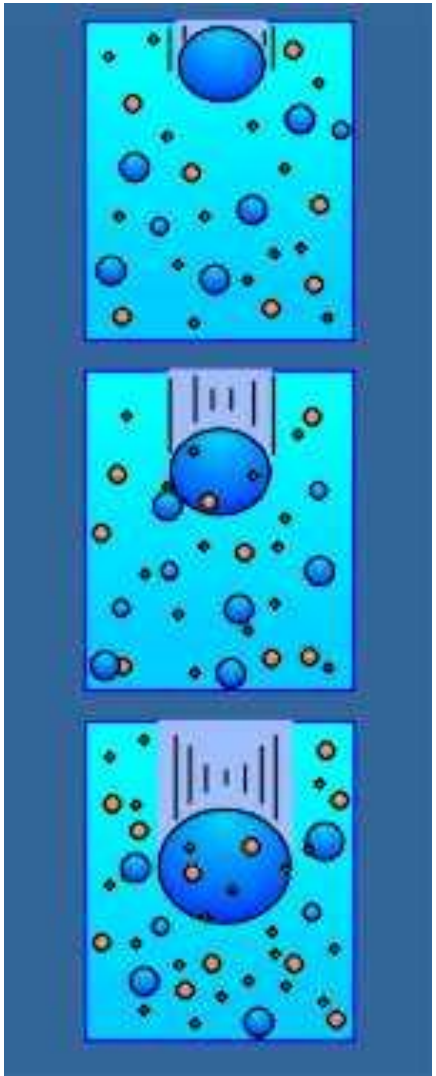
http://dionysos.mpch-mainz.mpg.de/smocc/home.htm

Rain

Raindrops form on cloud condensation nuclei (CCN) aerosols (~ 0.2 µm diameter)

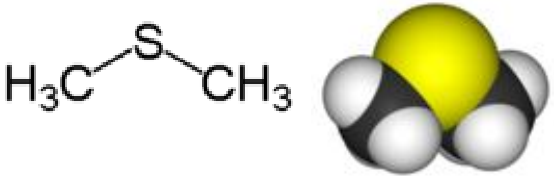
Raindrops:

- Collide and scavenge other atmospheric material
- Dissolve material from scavenged solid aerosols
- Absorb atmospheric gases



Gas-to-particle conversion

SO_4^{2-} (sulfate) aerosol is produced from atmospheric oxidation of *dimethylsulfide (DMS*), which is produced by plankton



Other sulfate aerosol sources include sulfur dioxide from combustion of coal and other fossil fuels ($SO_2 \rightarrow SO_4^{2-}$)

Oxides of nitrogen from combustion are converted to nitrate aerosol ($NO_x \rightarrow NO_3^{2-}$)

Oxidation of SO_2 to H_2SO_4 and partial neutralization with NH_3 yields *hydroscopic* particles rich in ammonium sulfate $(NH_4)_2SO_4$

Atmospheric Deposition

Transfer of material from the atm to the earth's surface

Wet deposition

Rainout: incorporation of material in clouds, requires nucleation of raindrops

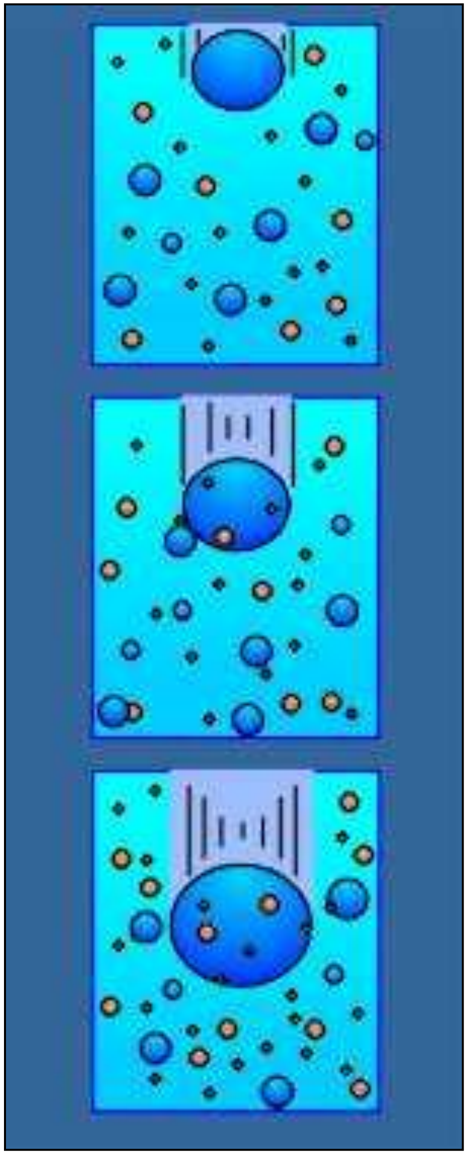
Washout: scavenging of material by rain as it falls

Inverse relationship between conc of component in atm and amount of rain, as continuing rain "cleanses" atmosphere

Smaller drop sizes have higher concentrations. Thus, *fog waters* can have very high concs of material; important for high elevation coastal plants

Scavenging ratio (s.r.) =

$$\frac{\text{Amount of material in rain (mg/L)} * \text{rain volume (L /m}^3\text{)}}{\text{Amount of material in air (mg/m}^3\text{)}}$$



Use rainvolume to make dimensionless ratio

Can calculate s.r. for each component from aerosol and rain measurements

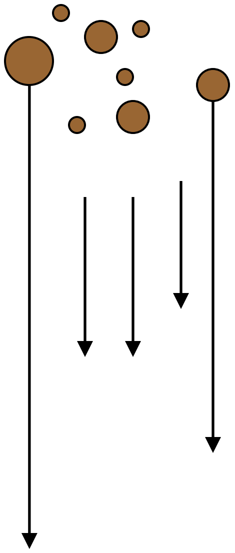
High s.r. for easily scavenged materials (*e.g.*, elements from large particles, soluble gases etc.)

Dust values range from 200 (Atlantic) to 1000 (Pacific)

Snowfall lowers s.r. (less efficient than rain)

Dry deposition

From gravitational settling of particles



Very important downwind of desert regions with *loess soils* (deposits of silt (2-64 µm diameter) that have been laid down by wind action)

Collect dry deposition with collectors that close during rain

Can get "local" contamination -- leads to overestimation of flux

Some particulate material is readily soluble in ground waters, so is important source of nutrients to soils -- especially in regions with low release rates of nutrients from soil weathering

Gas absorption

Direct absorption of N and S gases by plants – important in humid regions (*e.g.*, Tennessee forest receives 75% of N input from absorption)

Problem with wet/ dry deposition measurements on vegetation

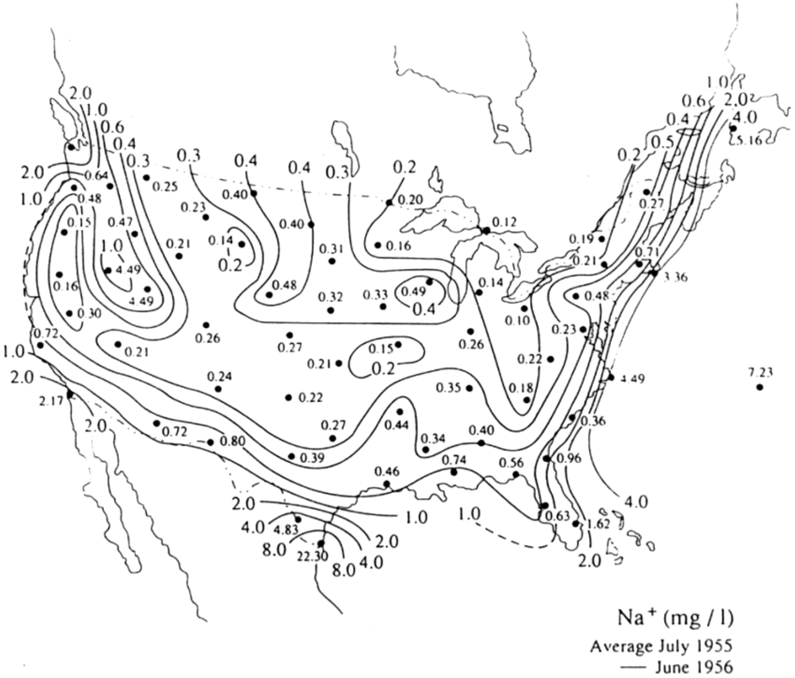
Hard to measure direct impaction of material moving *horizontally* onto leaf surfaces – or upwards onto the lower surfaces of leaves

Regional patterns of deposition

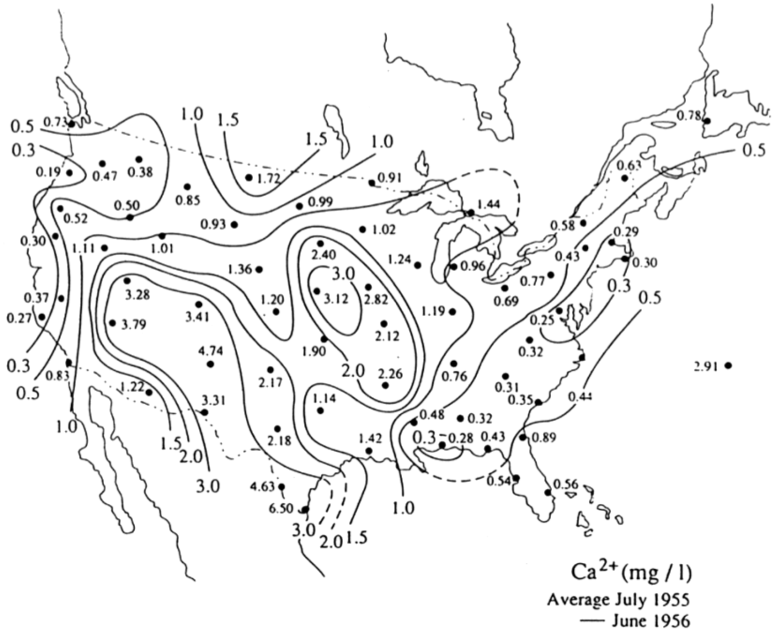
Reflect relative importance of sources

Coastal regions have high sea-salt components (*e.g.*, Na, Cl)

Arid regions have high soil components (*e.g.*, Ca, Fe)



Na^+ (mg / l)
Average July 1955
— June 1956



Ca^{2+} (mg / l)
Average July 1955
— June 1956

Precipitation

Regions downwind of pollution sources have high SO_4^{2-}, low pH etc.

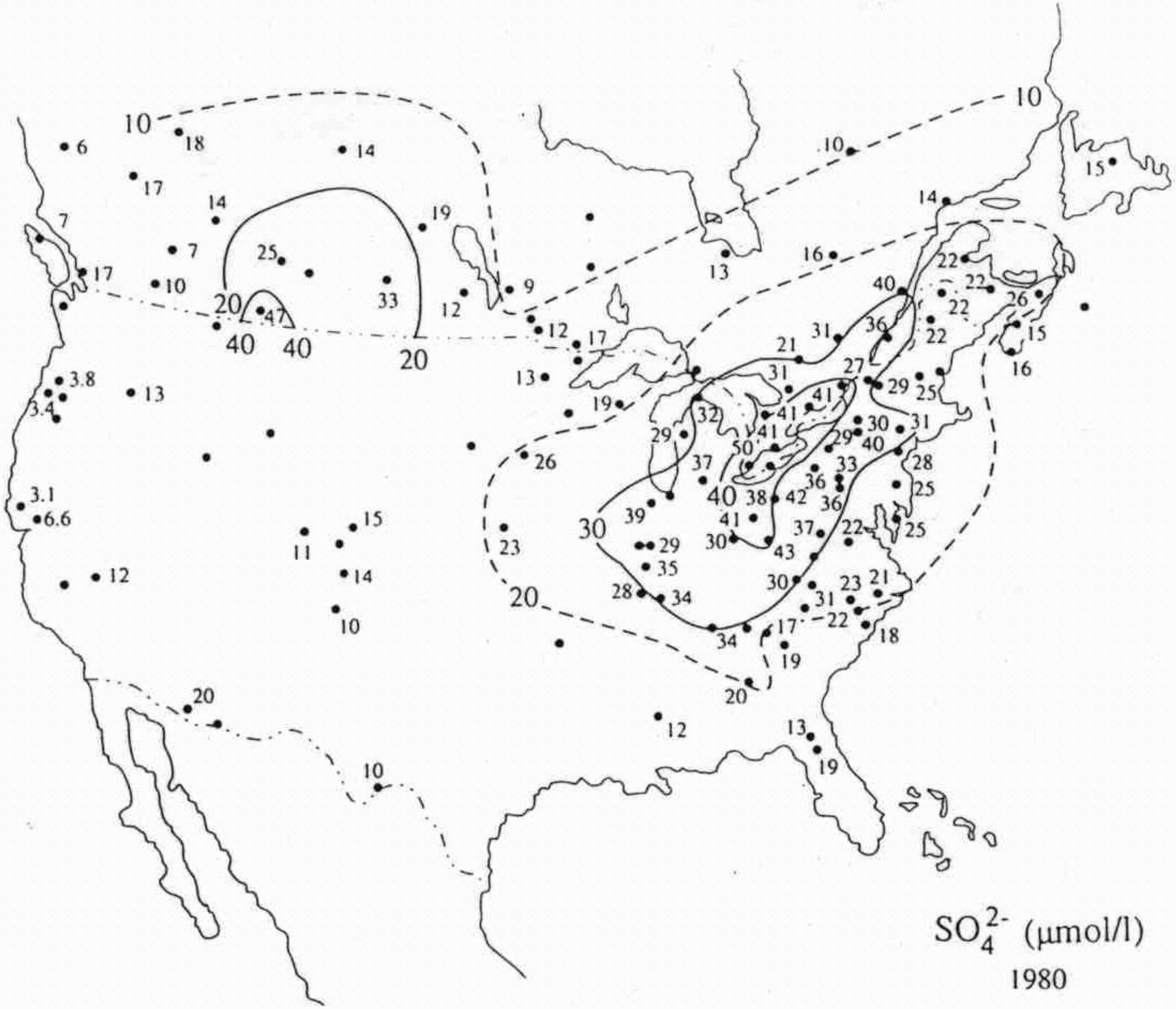
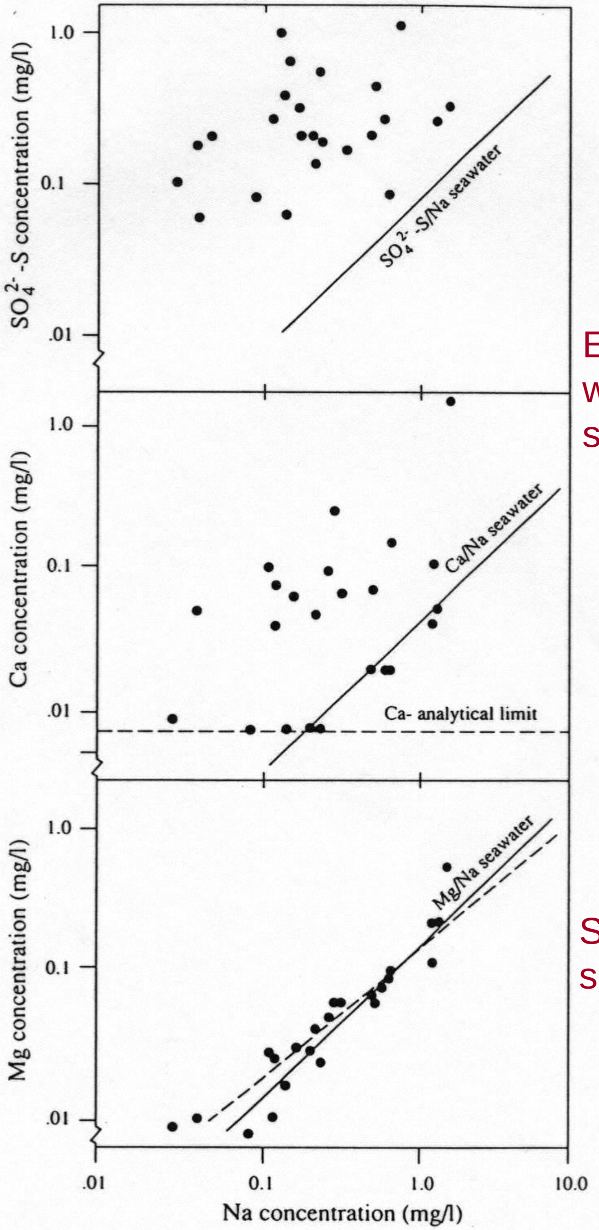


Figure 3.15 *(continued)*

Figure 3.15 Geographic pattern in the concentration of some major constituents in U.S. precipitation. Na and Cl are from Junge and Werby (1958) and SO_4 is modified from Barrie and Hales (1984).

Can use chemical ratios in rain to identify sources

E.g., ratio of element to Na is used to identify sea-salt component of element when you have multiple sources



Enriched wrt seawater

Same as seawater

Figure 3.16 Concentrations of SO_4, Ca, and Mg in wetfall precipitation near Santa Barbara, California, plotted as a logarithmic function of Na concentrations in the same collections (Schlesinger et al. 1982). The solid line represents the ratio of these ions to Na in seawater. Ca and SO_4 are enriched in wetfall relative to seawater, whereas Mg shows a correlation (dashed) that is not significantly different from the ratio expected in seawater.

Al and Fe can be used to calculate dust components and, by ratio, other elements from dust sources

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Correlation of H^+ and SO_4^{2-} downwind of pollution sources – due to oxidation of anthropogenic SO_2. NO_3 also can contribute to acidity.

Note: equilibrium pH for rain in contact with current atm CO_2 levels = 5.6

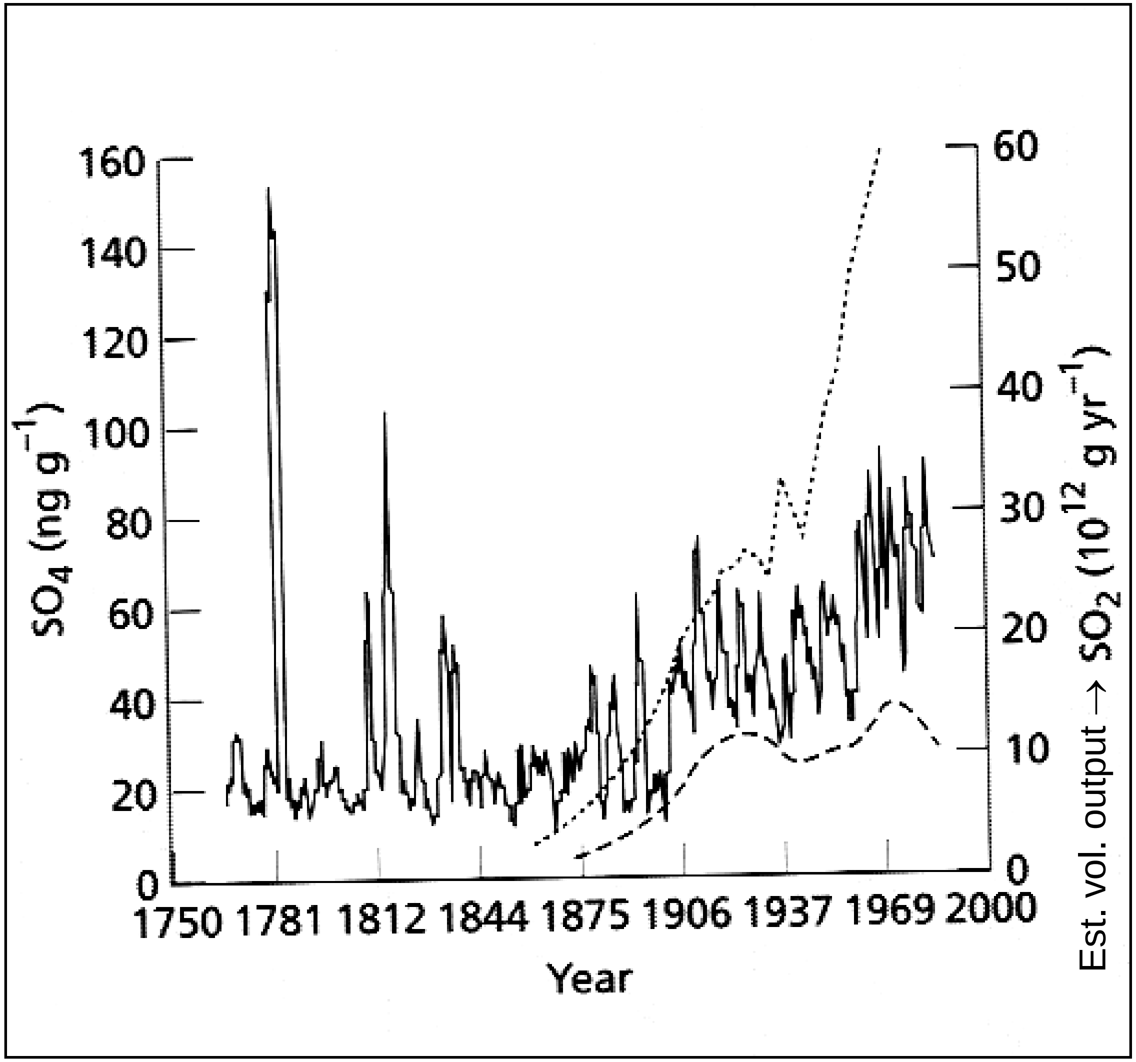
Rainfall pH buffered by NH_4^+ and Ca^{2+} from vegetation and soils

In rain: $H^+ = [NO_3^- + 2SO_4^{2-}] – [NH_4^+ + 2Ca^{2+}]$

Globally, 22% of rain acidity is neutralized; greater in Southern Hemisphere (less pollution)

SO_4^{2-} and NO_3^- recorded in Greenland ice-cores increased 3 - 4 times since 18th century:

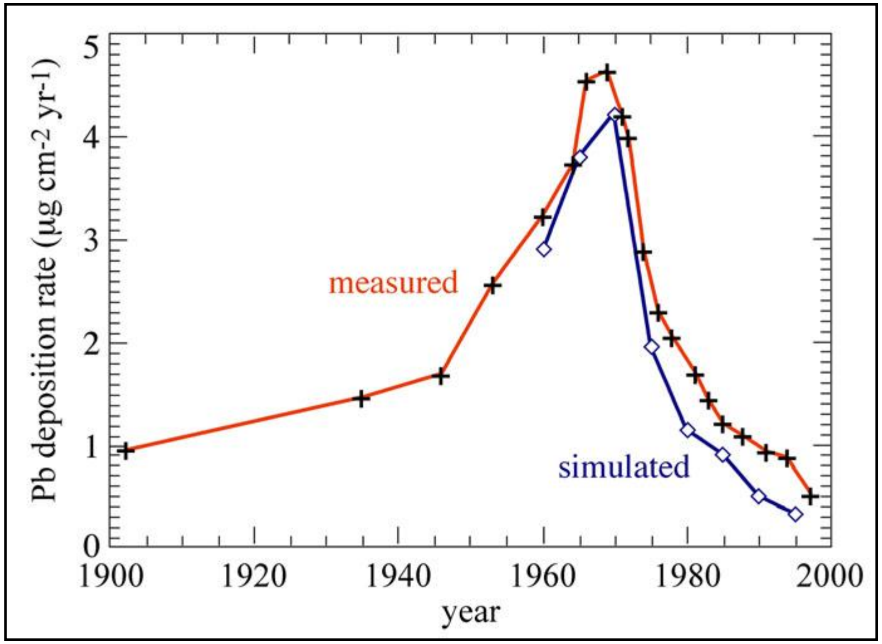
Sulfate levels in the Greenland ice cores show recent increases matching both global and US increases in SO_2 output:



Deposition of SO_4^{2-} in eastern US is 2 – 16 times background rates

Other trace element increases (*e.g.*, Pb, Hg) seen in lake sediments from anthropogenic activity

Recent controls on emissions may reduce some materials (*e.g.*, Pb); seen in Sargasso Sea (north-central Atlantic), and Danish peat:



Conflicting effects – *e.g.*, N deposition can increase fertility, but increased H^+ can lead to deficiencies in other nutrients

Atmospheric Models

Model classification

1-D models consider vertical column (Z) of atmospheric processes, but apply same values to all of the Earth surface (*i.e.*, no variation in X or Y)

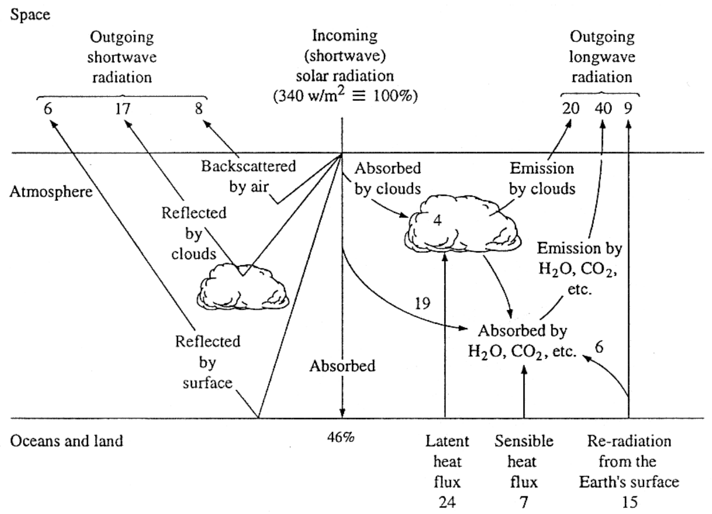


Figure 3.2 The radiation budget for Earth, showing the proportional fate of the energy that the Earth receives from the Sun. Each year, the Earth receives about 340 W/m² of radiation, mostly in short wavelengths. About a third of this radiation is reflected back to space, and the remainder is absorbed by the atmosphere (23%) or the surface (46%). Long-wave (infrared) radiation is emitted from the Earth's surface, some of which is absorbed by atmospheric gases, warming the atmosphere (the greenhouse effect). The atmosphere emits long-wave radiation, so that the total energy received is balanced by the total energy emitted from the planet. Modified from MacCracken (1985).

2-D models allow variation in parameters or processes along one horizontal scale (X or Y) (*e.g.*, latitude)

3-D models (General Circulation Models, GCMs) allow vertical and horizontal exchange of properties along X and Y directions.

GCMs can show fates of parcels of air

Can include chemical & physical processes

The large number of chemical rxns can make them very complex

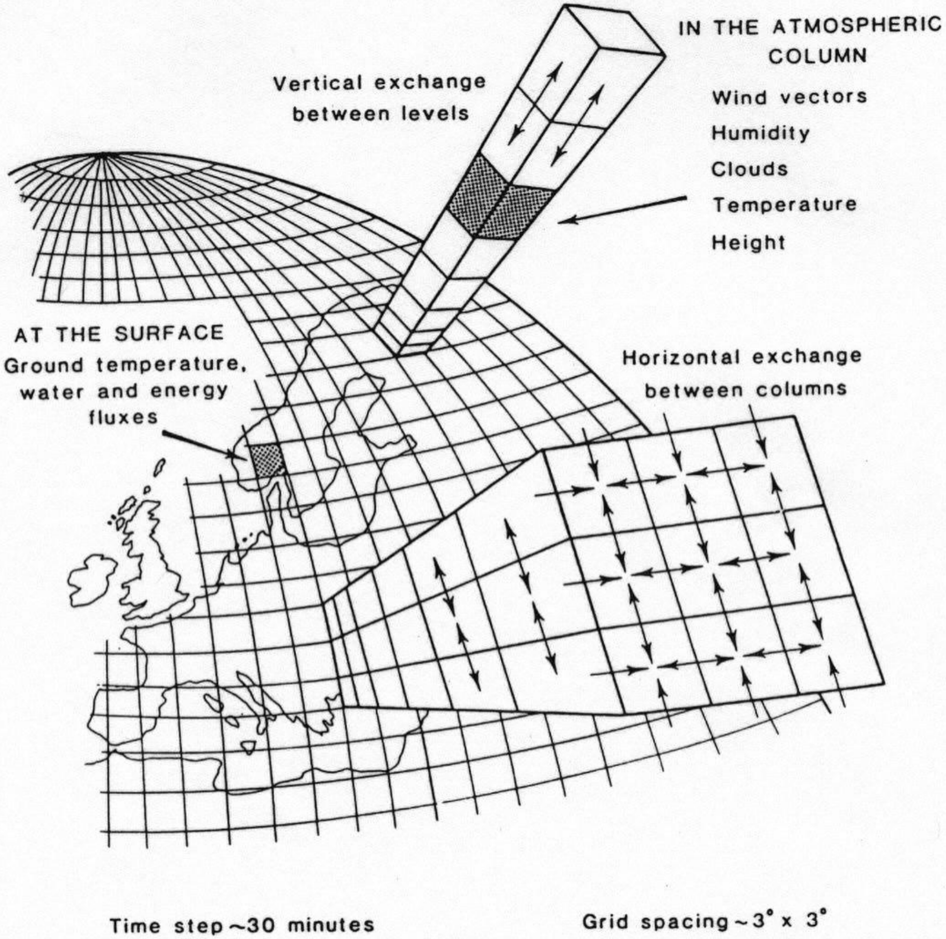


Figure 3.13 Conceptual structure of a dynamic, three-dimensional general circulation model for the Earth's atmosphere, indicating the variables that must be included for a global model to function properly. From Henderson-Sellers and McGuffie (1987).

Limiting factors on model accuracy

- Initial-condition data
- Computational speed – for complex models, one day of model can take nearly a day to calculate!

Consensus results

Nearly all models predict atm warming of 1.5 - 5.5˚C from increased IR absorption by greenhouse gases

Greatest warming at poles, where IR net loss is greatest

Warming of ocean will absorb heat → increased water evaporation → increased greenhouse effect (positive feedback)

Expect changes in next century greater than in last 2 Myr

Currently hard to see global effects from satellite data, but regional temperature increases appear

Can see effect from surface-based measurements:

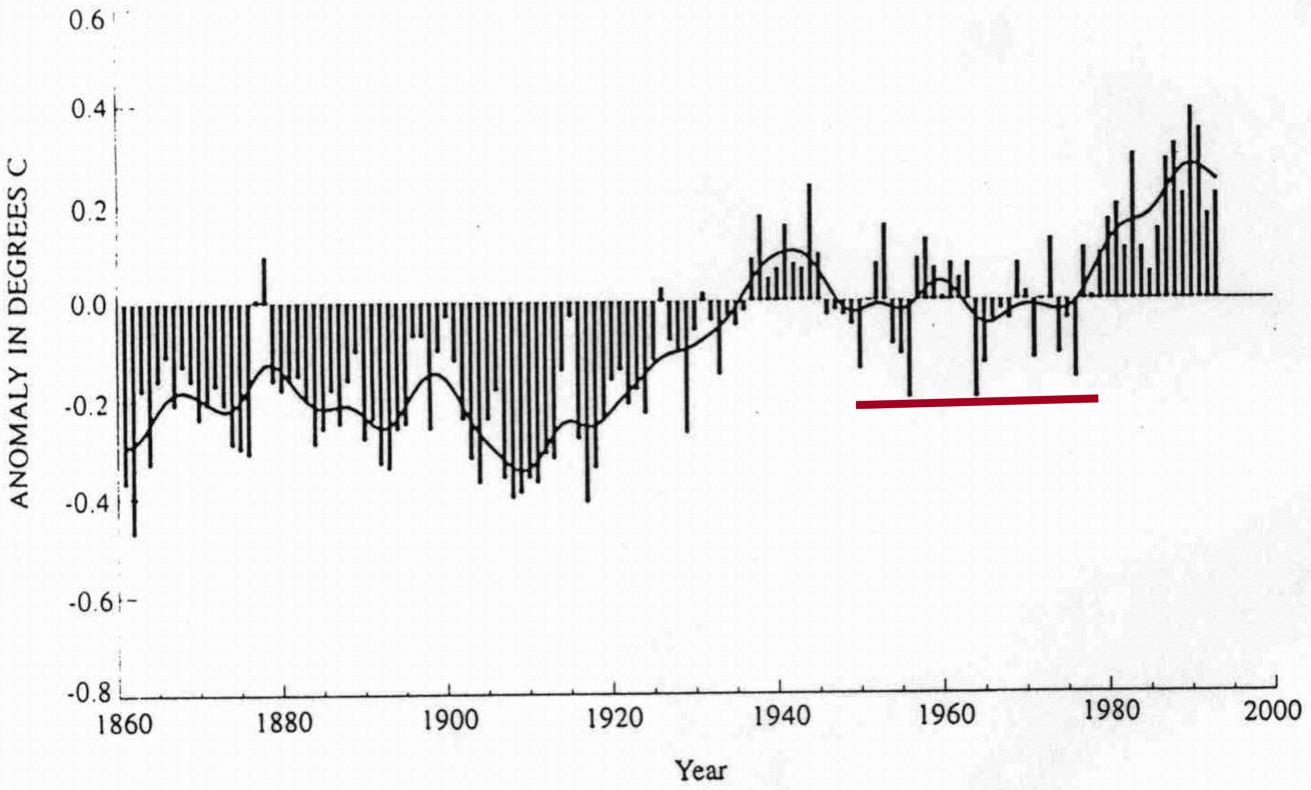


Figure 3.14 Mean global temperature, derived from measurements made on land and at sea, for the period 1861–1993, shown as deviations from the mean in the interval from 1951 to 1980. From Hadley Center for Climate Prediction and Research, United Kingdom.

Cooling effects due to aerosols

Increasing SO_4^{2-} and dust aerosols reflect more incoming radiation

Also increase cloud formation through CCN effect

Clouds increase reflectivity, slowing increase in warming

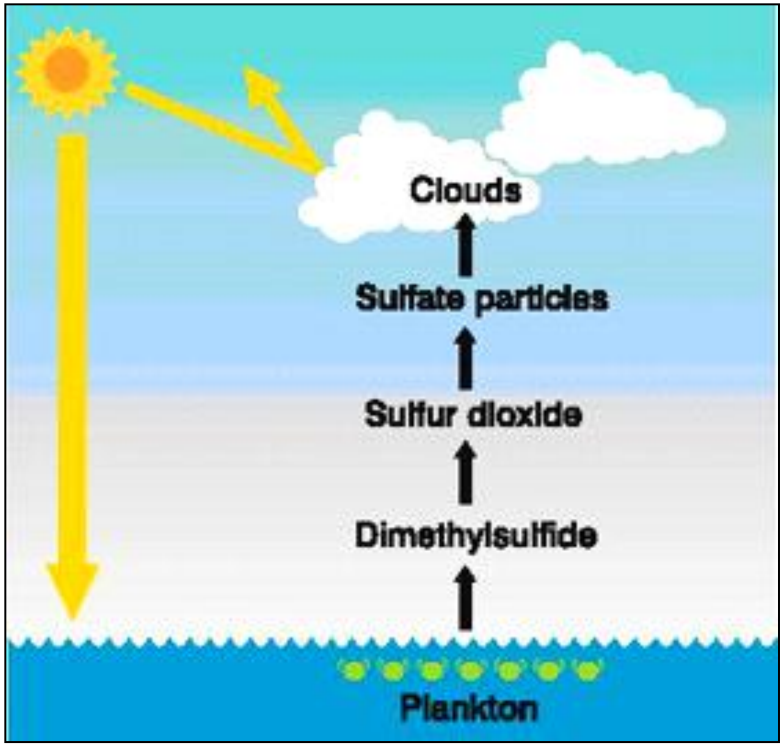
Thus, models now include aerosols → better T predictions

Radiation balance:

Incoming	340 W/m^2
Natural greenhouse trapping	153 W/m^2
Anthropogenic greenhouse	2.1 W/m^2
Increased clouds	-0.3 W/m^2

CLAW hypothesis

- Named for the four scientists who formulated it - **C**harlson, **L**ovelock, **A**ndreae and **W**arren (Charlson *et al.*, 1987)



www.whoi.edu/science/

- In the following decade, was the subject of >700 scientific papers describing the biogeochemistry of DMS, its precursors, and the connection to earth's climate

- Now have evidence that some of the steps within the CLAW hypothesis are correct – but we still don't know whether the system really operates as a negative feedback loop

- One hint: last glacial max had higher aerosols and lower CO_2 -- maybe connected through enhanced Fe inputs to ocean, and greater oceanic photosynthesis

The use and abuse of climate models

Kevin E. Trenberth

Projections of future climate change depend largely on the results of computer models. Such models are becoming increasingly sophisticated, but they do not offer the certainties that policy-makers would like.

Humankind is performing a great geophysical experiment[1]. By modifying the Earth's environment in various ways, we are changing the climate. The extent and the rate of these changes are unclear, as is what (if anything) should be done about them, but that the experiment is underway is not in doubt. The environmental changes of most relevance are in land use (farming, building cities), storage and use of water (dams, reservoirs, irrigation), generation of heat, and — most notably — the burning of fossil fuels.

In particular, fossil-fuel combustion pollutes the atmosphere and alters the balance of radiation on Earth through both visible particulate pollution (called aerosols), and gases that change the composition of the atmosphere. These are known as greenhouse gases because they are relatively transparent to incoming solar radiation, but absorb and re-emit outgoing infrared radiation, thus creating a blanketing effect which results in warming. For example, as a consequence of human activities, carbon dioxide concentrations in the global atmosphere[2–4] have increased by about 30 per cent over pre-industrial values. Global warming and associated climate change is expected as a result, and the global mean temperatures have indeed risen over the past hundred years[5] (Fig. 1).

If this experiment turns out badly — however that is defined — we cannot undo it. We cannot even abruptly turn it off, because too many of the things we are doing now have long-term ramifications. For instance, carbon dioxide has an atmospheric lifetime of over a century[5] and simply stopping increases in emissions would still result in increases in atmospheric concentrations for many decades. The only way to reverse those trends is to reduce emissions to well below current levels[4,5]. Moreover, changes underway in the oceans would endure, because of the oceans' huge heat capacity.

If we had two planet Earths, identical in every respect except that the residents of one adopted measures to avoid polluting the atmosphere while residents of the other did not, we could see how the climates of the two planets would diverge, and what the consequences would be. But we don't and we can't. Instead we have to do the next best thing — try to understand the climate system well enough to build a good model of the planet Earth system and use this model to perform the experiments. We can indeed construct a miniaturized physical model of the Earth–Sun system; but we cannot readily include the effects of gravity, and the rich complexity of the atmosphere and oceans. The alternative is to build a virtual model of the Earth in a computer.

The models

These computer models are based upon physical laws represented by mathematical equations and expressions that are solved using numerical methods as applied to a three-dimensional grid over the globe. The most complete versions are referred to as 'Earth system models' and those which deal exclusively with climate are called 'climate models' or, if they are very comprehensive, 'climate system models'[6].

But how useful are these models in making projections of future climate? Opinion is polarized. At one extreme are those who take the model results as gospel; at the other are those who denigrate such results simply because they distrust models, or on the grounds that the model performance is obviously wrong in some respect or that a process is not adequately included. The truth lies in between. All models are of course wrong because, by design, they depict a simplified view of the system being modelled. Nevertheless, many — but not all — models are very useful.

A full climate system model[6] should deal with all of the physical, chemical and biological processes that occur in nature and the interactions among the components of the climate system (Fig. 2, overleaf). A major component of them is the so-called atmospheric general circulation models (AGCMs); these are designed to simulate the detailed evolution of weather systems and weather phenomena, as well as the physical and dynamical processes involved. For AGCMs, typical resolutions for climate simulations are about 250 km in the horizontal direction and 1 km in the vertical. These models are widely used and tested every day in making weather forecasts, although with finer resolution, and have predictive value out to about ten days ahead[7].

The theoretical limit to the predictability of weather is about two weeks, which stems from the phenomenon of chaos — small uncertainties in the analysis of current weather conditions rapidly grow and eventually become large enough to make the forecast worthless[8]. This essentially random-error component is overcome in climate forecasts by predicting only the statistics of the weather (that is, the climate). Accordingly, systematic influences of changing conditions in the ocean, sea ice, land surface, solar radiation or other factors are reflected in atmospheric variations on various time scales. The most obvious example is the climate change with the seasons. An ensemble of several climatic simulations, each of which begins from somewhat different starting conditions, can be used to establish which climatic features are reproducible in the simulations and thus are predictable with the model.

These features constitute the climate signal, while those which are not reproducible can be considered weather-related climate noise. Climate predictability is a function of spatial scales. Natural atmospheric variability is enormous on small scales and most effects on climate (forcings), such as from increases in greenhouse gases, are predominantly global. So the noise level of natural variability will mask a climate signal more as smaller regions are considered[5].

Model errors

The latest coupled atmosphere–ocean–land–sea-ice models provide very good simulations of average climate conditions and

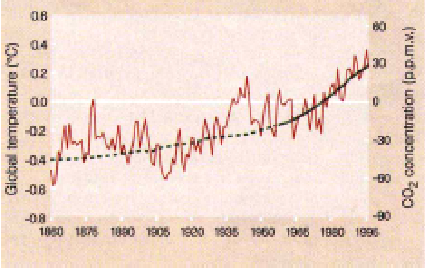


Figure 1 **Estimated changes in annual global mean temperatures[5] (red) and carbon dioxide (green) over the past 137 years relative to a 1961–90 base period. Earlier values for carbon dioxide are from ice cores[3] (dashed line), and for 1957 to 1995 from direct measurements made at Mauna Loa, Hawaii[2]. The scale for carbon dioxide is in parts per million by volume (p.p.m.v.) relative to a mean of 333.7 p.p.m.v.**

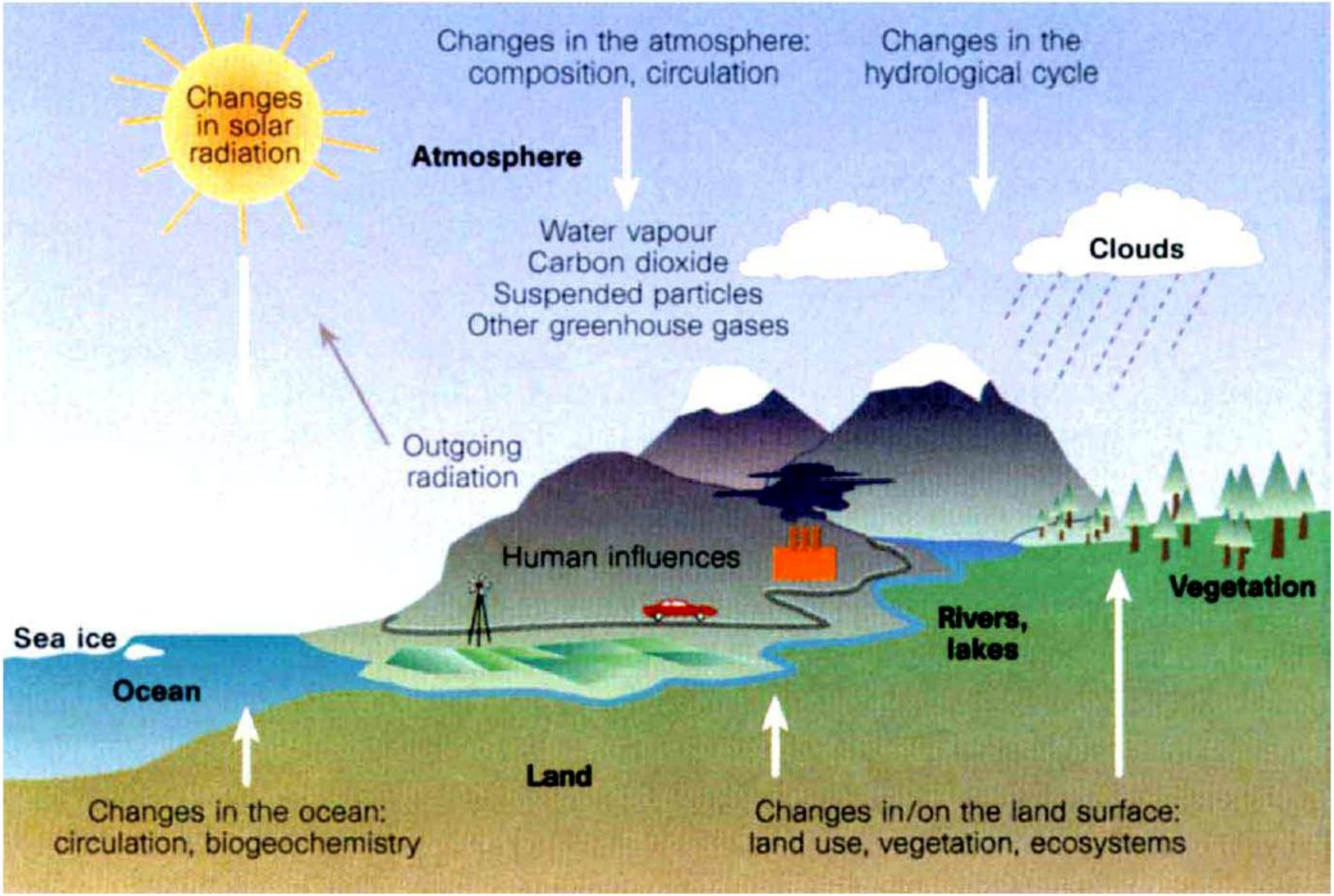


Figure 2 **Components of the global climate system — atmosphere, ocean, sea ice, land surface, surface hydrology and biosphere — their processes and interactions, and some aspects that may change.**

All of these factors need to be included in global climate models

The details of a model can greatly affect the results!

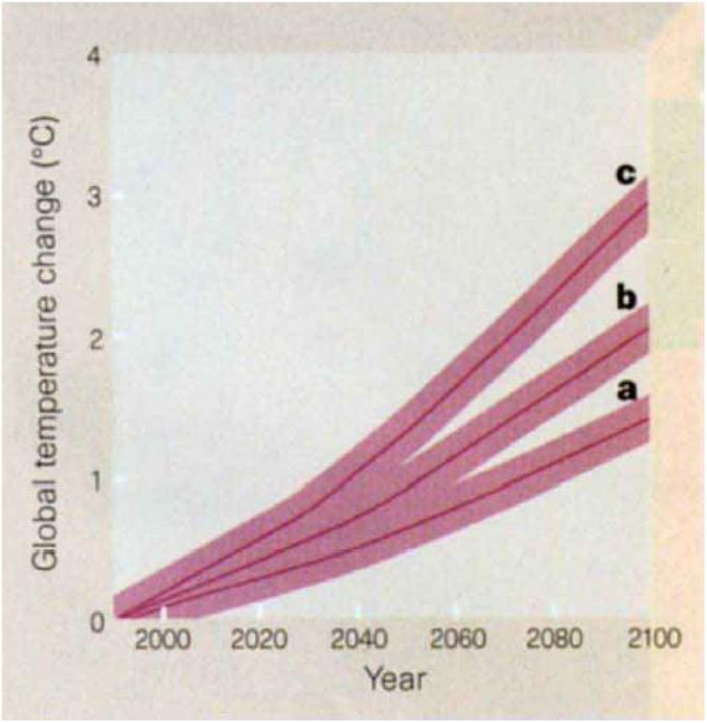


Figure 3 **Projected changes in global mean temperature from 1990 to 2100, corresponding to an emissions estimate[12] involving increases in both carbon dioxide from 350 to 700 p.p.m.v. and in sulphate aerosol (a 'mid-range scenario'). The three curves show the average changes and the scatter (shading) from natural variability corresponding to models with low** (a)**, best estimate** (b) **and high sensitivity to change** (c)**.**

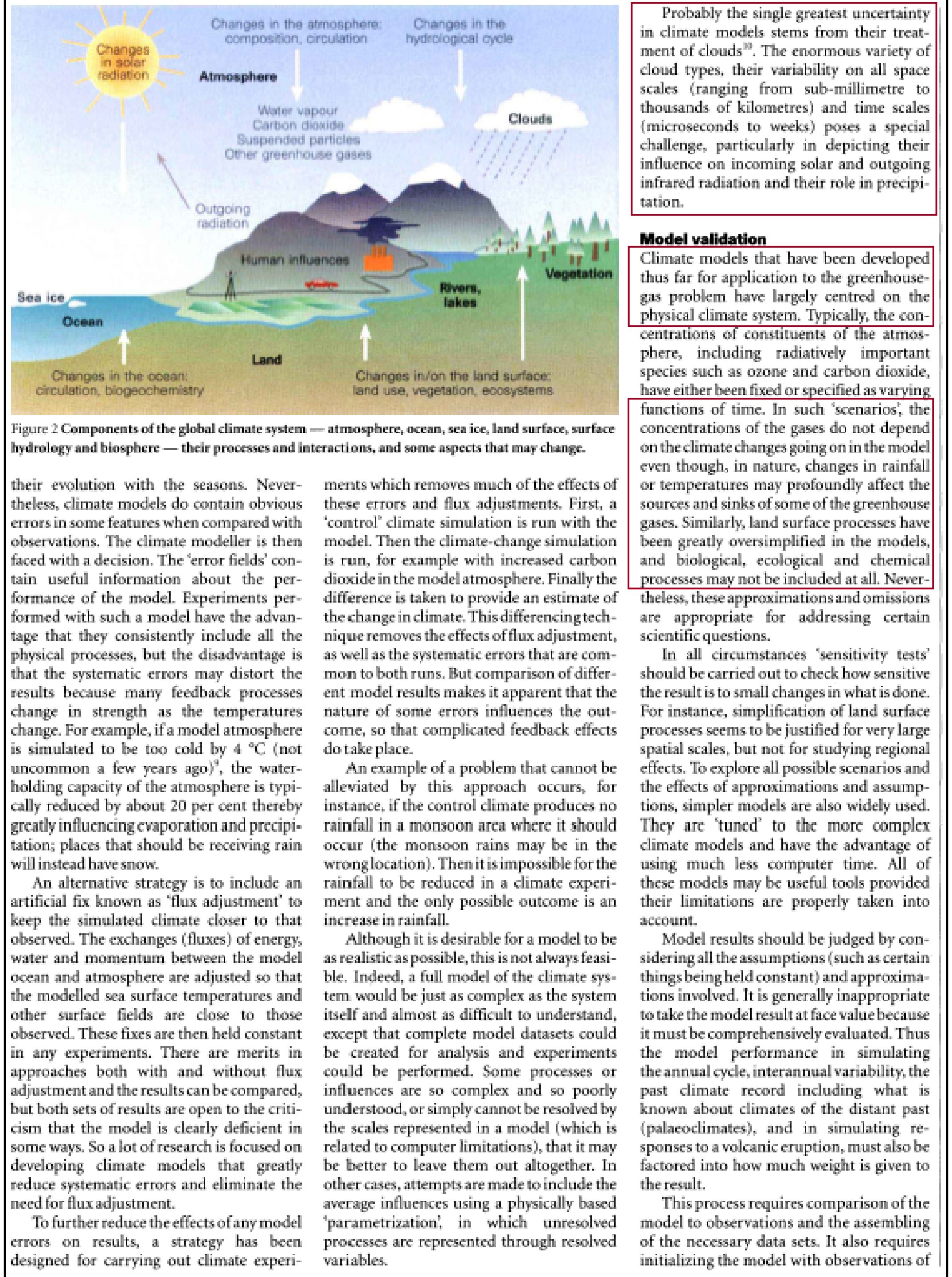


Figure 2 **Components of the global climate system — atmosphere, ocean, sea ice, land surface, surface hydrology and biosphere — their processes and interactions, and some aspects that may change.**

their evolution with the seasons. Nevertheless, climate models do contain obvious errors in some features when compared with observations. The climate modeller is then faced with a decision. The 'error fields' contain useful information about the performance of the model. Experiments performed with such a model have the advantage that they consistently include all the physical processes, but the disadvantage is that the systematic errors may distort the results because many feedback processes change in strength as the temperatures change. For example, if a model atmosphere is simulated to be too cold by 4 °C (not uncommon a few years ago)[9], the water-holding capacity of the atmosphere is typically reduced by about 20 per cent thereby greatly influencing evaporation and precipitation; places that should be receiving rain will instead have snow.

An alternative strategy is to include an artificial fix known as 'flux adjustment' to keep the simulated climate closer to that observed. The exchanges (fluxes) of energy, water and momentum between the model ocean and atmosphere are adjusted so that the modelled sea surface temperatures and other surface fields are close to those observed. These fixes are then held constant in any experiments. There are merits in approaches both with and without flux adjustment and the results can be compared, but both sets of results are open to the criticism that the model is clearly deficient in some ways. So a lot of research is focused on developing climate models that greatly reduce systematic errors and eliminate the need for flux adjustment.

To further reduce the effects of any model errors on results, a strategy has been designed for carrying out climate experiments which removes much of the effects of these errors and flux adjustments. First, a 'control' climate simulation is run with the model. Then the climate-change simulation is run, for example with increased carbon dioxide in the model atmosphere. Finally the difference is taken to provide an estimate of the change in climate. This differencing technique removes the effects of flux adjustment, as well as the systematic errors that are common to both runs. But comparison of different model results makes it apparent that the nature of some errors influences the outcome, so that complicated feedback effects do take place.

An example of a problem that cannot be alleviated by this approach occurs, for instance, if the control climate produces no rainfall in a monsoon area where it should occur (the monsoon rains may be in the wrong location). Then it is impossible for the rainfall to be reduced in a climate experiment and the only possible outcome is an increase in rainfall.

Although it is desirable for a model to be as realistic as possible, this is not always feasible. Indeed, a full model of the climate system would be just as complex as the system itself and almost as difficult to understand, except that complete model datasets could be created for analysis and experiments could be performed. Some processes or influences are so complex and so poorly understood, or simply cannot be resolved by the scales represented in a model (which is related to computer limitations), that it may be better to leave them out altogether. In other cases, attempts are made to include the average influences using a physically based 'parametrization', in which unresolved processes are represented through resolved variables.

Probably the single greatest uncertainty in climate models stems from their treatment of clouds[10]. The enormous variety of cloud types, their variability on all space scales (ranging from sub-millimetre to thousands of kilometres) and time scales (microseconds to weeks) poses a special challenge, particularly in depicting their influence on incoming solar and outgoing infrared radiation and their role in precipitation.

Model validation

Climate models that have been developed thus far for application to the greenhouse-gas problem have largely centred on the physical climate system. Typically, the concentrations of constituents of the atmosphere, including radiatively important species such as ozone and carbon dioxide, have either been fixed or specified as varying functions of time. In such 'scenarios', the concentrations of the gases do not depend on the climate changes going on in the model even though, in nature, changes in rainfall or temperatures may profoundly affect the sources and sinks of some of the greenhouse gases. Similarly, land surface processes have been greatly oversimplified in the models, and biological, ecological and chemical processes may not be included at all. Nevertheless, these approximations and omissions are appropriate for addressing certain scientific questions.

In all circumstances 'sensitivity tests' should be carried out to check how sensitive the result is to small changes in what is done. For instance, simplification of land surface processes seems to be justified for very large spatial scales, but not for studying regional effects. To explore all possible scenarios and the effects of approximations and assumptions, simpler models are also widely used. They are 'tuned' to the more complex climate models and have the advantage of using much less computer time. All of these models may be useful tools provided their limitations are properly taken into account.

Model results should be judged by considering all the assumptions (such as certain things being held constant) and approximations involved. It is generally inappropriate to take the model result at face value because it must be comprehensively evaluated. Thus the model performance in simulating the annual cycle, interannual variability, the past climate record including what is known about climates of the distant past (palaeoclimates), and in simulating responses to a volcanic eruption, must also be factored into how much weight is given to the result.

This process requires comparison of the model to observations and the assembling of the necessary data sets. It also requires initializing the model with observations of

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