ATMOSPHERIC CHEMISTRY AND CLIMATE IN THE ANTHROPOCENE*

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I will speak about the Anthropocene, which is a new geological era created by mankind in the last 200 years or so, and I will also show you some results of the geoengineering exercises that we have been doing, basically to cool down the earth from too much heat caused by greenhouse gases.

Since the beginning of the 19th century we can agree that mankind has really opened a new geological era. Normally it is claimed we are in the Holocene, but we are no longer in the Holocene, we are in the Anthropocene because, in many ways, we determine the climate of the earth, its atmospheric chemistry and conditions at the surface. I shall give you some examples.

During the past three centuries human population increased by a factor of 10 and, in the last century which has just ended, by a factor of 4. Cattle population increased by 1400 million, that is, one cow per average family. Cattle produce methane, which we are interested in since methane is a greenhouse gas and also determines much of the background chemistry of the atmosphere. Urbanisation has grown more than tenfold in the past century and almost half of the people now live in cities, in megacities, and this tendency is increasing, especially in the developing countries. Industrial output increased 40 times during the past century and energy use 16 times, and almost 50% of the land surface has been transformed by human activity.

Water use increased by nine fold during the past century, to about 800 cubic metres per capita, most of which is used for irrigation, 25% for industry and 10% for households. To give some examples of the use of water resources, it takes about 20,000 litres of water to grow 1 Kg of coffee, 11,000 litres of water to make a quarter pounder and 5,000 litres of water to make 1 Kg of cheese: quite impressive, so no wonder we are running dry.

^{*} This is a transcript of the author's lecture during the Plenary Session, which the PAS is publishing as is, without the author's corrections.

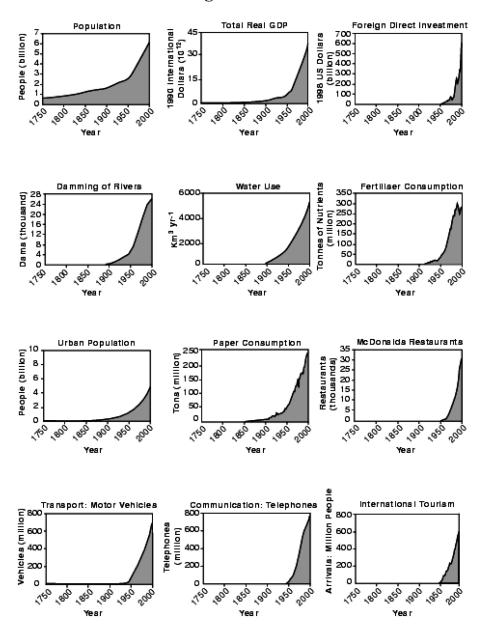
Another example of human activities is the appropriation of terrestrial net primary productivity: it looks like mankind is using about 30% of the natural resources available in terrestrial net primary productivity. Fish cash increased 40 times, the release of sulfur dioxide to the atmosphere, which only 2 decades ago was 160 Tg/year (a teragram is 10¹² grams or 1 million ton), is fortunately now down to 110 Tg/year. There has been an improvement, and that is because sulfur emissions had caused major problems, for instance acid rain, bad effects on health, poor visibility, and also have an impact on cloud formation and sulfate aerosol formation. Release of NO to the atmosphere from fossil fuel and biomass burning is larger than its natural inputs, causing high surface ozone levels over extensive regions of the globe.

However, several climatically important 'greenhouse gases' have substantially increased in the atmosphere: carbon dioxide by more than 30% and methane by more than 100%. Most of these changes have actually taken place or picked up since the end of the last world war, so this is what we call the *Great Acceleration* (Fig. 1): for instance, population increase, total real gross domestic product, foreign direct trade, the damming of rivers, which is a major activity of mankind and, also, the growth of McDonald's around the world which, of course, has to do with methane release and the involvement of cows. I could mention many more examples but these may suffice.

Humanity is also responsible for the presence of many toxic substances in the environment, even some that are not toxic at all but that have, nevertheless, led to the ozone hole, and those are, of course, the chlorofluorocarbon gases. CFC gases are very inert in the troposphere, are destroyed by ultraviolet radiation above about 25 km in the atmosphere and then give rise to chlorine atoms that break down in the stratosphere. This was proposed and hypothesised for the first time by Mario Molina, who is in the audience. They also cause UV-B radiation and skin cancer.

We also have species extinction. The natural extinction rate of species was roughly 1 species per million species per year: it is now about a factor a thousand time larger, so the average life span of species in the atmosphere is close to one thousand times shorter than in pre-industrial, pre-Anthropocene conditions.

Regarding erosion, we are experiencing 15 times the natural erosion rate as a result of human activities, man-caused erosion, crop tillage, land conversion for grazing, and construction. So, at the current rate, anthropogenic soil erosion would fill the Grand Canyon in about 50 years. We are disturbing the nitrogen cycle: here you can see (Fig. 2, see page 86), as a function of time, the natural nitrogen fixation rate by leguminous plants,



The great acceleration

Figure 1.

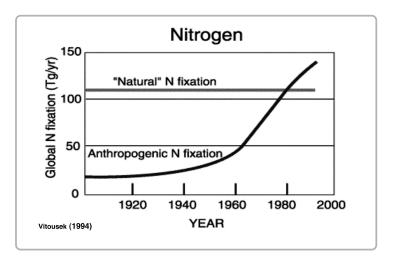


Figure 2.

and also by lightning, and you can also see the input by anthropogenic activities. Since about 1980 the anthropogenic nitrogen fixation, mainly fertiliser nitrogen, has been bypassing the natural N fixation rate, and that has major consequences for the emissions of nitrous oxide in the atmosphere. I will come back to that. It is amazing that anthropogenic nitrogen fixation is growing, although when we really look at what ends up in the mouths of people, that is only in the order of 10%, so 90% is sort of wasted. It really is a pity that agriculture is so inefficient in using its nitrogen.

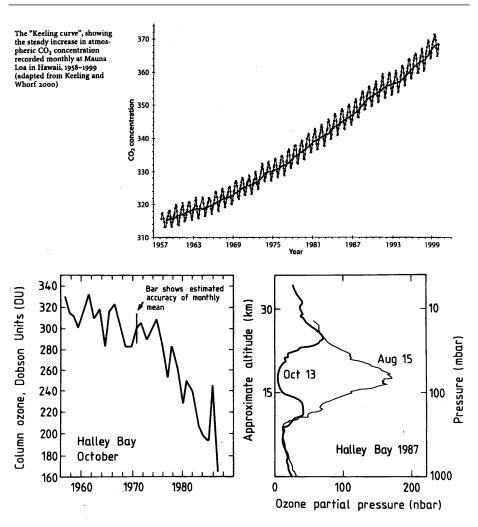
The composition of the atmosphere is dominated, as you all know, by nitrogen, oxygen, argon; we do not have to worry that we will run out of those, we leave the study of those compounds mainly to geologists. Atmospheric chemists and climate researchers nowadays are much more interested in the minor constituents in the atmosphere, starting with carbon dioxide, of which we now have 380 ppm (the numbers in the figure are a little outdated). It is growing by about 0.4% per year, it is, of course, the major greenhouse gas and, of course, it is involved in the biosphere and photosynthesis. It does not play a major role in the chemistry of the atmosphere. There we go down to gases with even lower concentrations, like methane, of which we have about 1.7 ppm in the atmosphere. This is double the amount of pre-industrial times, it has been growing quite considerably but, at the moment, is at a standstill, and I will briefly come

back to that. Ozone is an extremely important trace gas, at ground level it has both positive and negative effects. The positive effect at ground level is that it promotes the production of hydroxil radicals that clean the atmosphere. The bad effect is that, if there is too much ozone in the surface air that we breathe, it is not very healthy for people. In the stratosphere we have seen a decline of ozone because of the use of chlorofluorocarbons, which I will briefly discuss. It is very variable: we have, on average, about 30 parts per billion of ozone in the troposphere, while in the stratosphere we can have in the order of 10 millionth of ozone.

Nitrous oxide is a by-product of the nitrogen cycle. We now have about 0.32 ppm of nitrous oxide in the atmosphere and it is growing by about 0.25% per year. Then we have CFC gases. They are no longer growing, they are actually going down now very very slowly in the atmosphere because there has been international agreement to stop their production so the ozone layer will slowly recover.

I will show you some viewgraphs of the major effects of human activities (Fig. 3, see page 88): in the upper figure you see a steady rise of the carbon dioxide amount in the atmosphere. These data go back to the end of the 1950s, by Dave Keeling, who unfortunately died last year but left this record behind. You can also see the seasonal variations in carbon dioxide, having to do with photosynthesis, for instance. Then you see the ozone hole pictures below and you can see, in the picture on the left, a very drastic depletion in total ozone, since the early 1970s, over Antarctica, especially in the springtime month of October. If you look at the right hand side of this picture you see that the ozone destruction (what is shown there is the ozone concentration as a function of altitude) is especially happening at an altitude where we normally have a maximum of ozone. Within two months that ozone maximum had collapsed into an ozone minimum.

This was something that had never been predicted, so we have an example of what Prof. Zichichi was talking about, the data had to be collected, and initially the observers did not believe that data because this was so unexpected, something like this happening at the other side of the world where the CFC gases were not at all injected into the atmosphere. It is basically radical reactions, chlorine atoms which enter into catalytic reactions destroying ozone. Hence, for each chlorine atom produced from the CFCs, you destroy up to 100,000 ozone molecules before the chlorine is removed from the atmosphere. This is quite shocking, mankind has created a chemical instability in the atmosphere by the use of CFC gases, which look so innocent because you can even breathe them at ground level and they will not harm you very much, but this is what they are doing in the stratosphere.





Here we have figures showing the greenhouse effect (Fig. 4, see page 89). I will be very brief here. The earth is supplied by the sun, on average, by about 340 W/m², let us call that 100 units: of these 100 units almost 30 are scattered back to space, that is, by reflection at cloud tops and also by scattering of particles in the atmosphere. I will come back to that. There is also some absorption in the atmosphere in clouds and also elsewhere in the stratosphere by

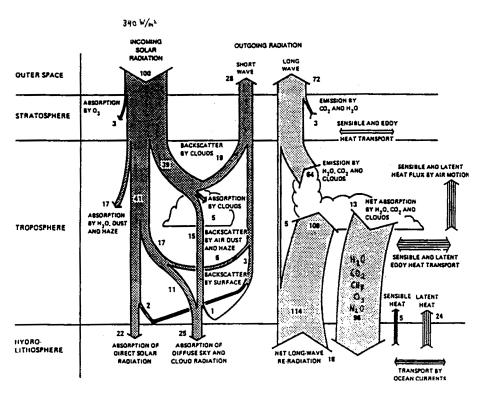


Figure 4.

water vapour, so what comes down to the earth's surface is about 47 units, the sum of 22 and 25. The earth has to get rid of this energy because, otherwise, in the space of a few weeks, it would start boiling around us. How is it done? It happens by the release of latent heat at the earth's surface, 24 units; 3 units sensible heat release, which added to 24 makes 27 units, and 47 units that we have to get rid of, so there are 18 units left. But now, in a very miraculous way, the earth and its atmosphere is taking care of the release of these 18 units, because it is not just emitting 18 units in the atmosphere, but it is emitting 140 units of which about 96 units are coming back, and that is due to the fact that the earth has the greenhouse gases in the first place, water vapour and carbon dioxide, but also methane, nitrous oxide, and ozone. The CFC gases are all contributing to this back flux of infrared radiation. So of the energy supplied by the sun, of the heat, there is about a factor of 6 recycling

of energy taking place, which, of course, makes life on earth possible at all, and which is due to the greenhouse gases, many of which have anthropogenic sources like CO_2 as I just mentioned.

The temperatures on earth really are rising (Fig. 5, see p. 268). You can see a steady rise in temperatures since 1970. The rise in temperature that we already had earlier may have had to do with natural variability of climate and maybe with some solar activity, but there is no doubt that nowadays, since 1970, we have had a steady rise in the temperatures in the atmosphere, as a global average, and it is continuing. Five or six of the warmest years were in the last decades, a clear sign that something is happening.

But the greenhouse gases are not the only factor that we have to consider. They heat the earth but there are also factors which lead to its cooling, and those are particles in the atmosphere (Fig. 6, see p. 269).

Now the uncertainties. The particles, many of which are released by human activities, cool the earth and also serve as cloud condensation nuclei, so they make the clouds more reflective to solar radiation, which has a cooling effect. The uncertainties, however, are very large, so we have to improve on that, but it will be a very difficult process to really estimate accurately what is the contribution of aerosol particles in the atmosphere. Many of these aerosol particles are produced by human activities, air pollution, and have a damaging function when you breathe them, so we really enter here into a dilemma because we want to get rid of these particles. due to their effects on health, but, by doing that, we increase the heating of climates, because the reflection of solar radiation to space is diminishing. This is a dilemma for policy-makers and, of course, us scientists and the general public. We have tried to estimate a little bit what the energy balance may be: the average amount of heat supplied by the sun to earth is 340 W/m^2 , the greenhouse forcing is 2.7 W/m^2 , the heating of the ocean is subtracting about 0.3 W/m², and, also because temperatures on earth have increased by about 0.6 to 0.7° in the atmosphere, we have an increased release of energy to space in the infrared by 1 W/m². But these factors combined, the 1.3 W/m², do not balance the 2.7 W/m² of heating, which means there is 1.4 W/m² left that the earth has to get rid of. Prof. Ramanathan and I, who did this analysis, think this is due to increased albedo effect, higher reflectivity of solar radiation by the clouds and, also, in general, the reflection of sunlight on the particles. Now it is interesting, if you improve the conditions, if you remove the particles from the atmosphere, meaning that the 1.4 W/m² becomes 0, basically, the net heating of the atmosphere will increase much more, about double as much compared to what is estimated here. It is instructive to note that the pure release of heat to the atmosphere by the burning of fossil fuels is only 0.025 W/m^2 , that is only 1% of the greenhouse forcing, so the real problem is the greenhouse gases, it is not the heat we put in the atmosphere, that has a minor effect. In fact, the heat released from earth is larger, 0.087 W/m^2 , than the heat released by the burning of fossil fuels.

So what are the effects? It is clear. The Intergovernmental Panel for Climate Change, under the auspices of the United Nations, brings out, every six years, its estimate of the situation, and what it said in 2001 (another report is due next year), is that there is new and stronger evidence that most of the warming observed over the last 50 years is attributable to human activities. The rise in temperatures during this coming century may be between 1.4 and 5.8°C, a very large uncertainty that has to do with uncertainty in science but also in the human behaviour in the future. How much fossil fuel are we going to burn in the future? All this leads to a major uncertainty.

These are big numbers, even the lower number here is quite substantial and will have effects on the earth's climate. It will cause the sea level to rise, estimated to maybe up to 80 cm, almost a centimetre per year in this century, since there are some signs that the upper limit of this range may be closer to the truth than the lower limit, there will be redistribution of precipitation, and, for instance, Italy and the Mediterranean regions will get substantially drier, which is something to worry about. Northern Africa I do not have to mention, lots of people are already moving away from Africa to Europe, enhanced risk of extreme weather, flooding, desertification, we had this very hot summer in 2003 and, again, this year is very odd from the point of view of meteorology. Too-rapid changes in temperatures will cause that the ecosystems cannot adapt to the situation.

What should we do about it? Well, in the first place, we should reduce emissions of greenhouse gases in the atmosphere, but that is easier said than done, because, to stabilise the amount of CO_2 in the atmosphere, we would have to reduce emissions by more than 60%, and that is not taking into account the growing contribution by developing countries, so will we ever be able to achieve the 60% or more reduction? One can be rather pessimistic, unfortunately. Methane I already mentioned: at the moment we do not see any increase in methane in the atmosphere for a while. That does not mean that it will not come back in the future, because with higher temperatures the permafrost regions in the northern latitudes, Canada and Russia, will thaw, which may lead to emissions of methane and carbon dioxide in the atmosphere and increased warming. A 70 to 80% reduction in the emissions of nitrous oxide would be required to stabilise its amount in the atmosphere, and this has to do with food production, nitrogen fertiliser, and I do not see that happening at all.

Fortunately, we have some success stories: CFC gases are no longer produced, only in very small amounts. But, nevertheless, you may have heard that this year was a very bad year for the ozone hole, the deepest ozone hole was just this year, despite the fact that these gases are very slowly disappearing from the atmosphere. But the activity of the CFC gases is also very much dependent on temperatures in the atmosphere. Clouding can only be activated if you have ice particles in the atmosphere, if you have this at higher latitudes, and this was a very cold year in the Antarctic, therefore causing very large ozone depletions. I am sure Mario Molina will go into detail.

In the reduction of the greenhouse gases, here we can see (Fig. 7, see p. 269) emissions in metric tons per capita per year, leading in North America, Oceania, Europe and you see all the developing countries here, which are hardly part of the game but they will be part of the game because they want to increase their standard of living. We should not believe that nature will help us out, that when temperatures go up then the greenhouse gases will go down cooling the earth, no, it is the opposite, when temperatures go up CO₂ also goes up at the same time, and methane goes up causing climate variability, because the Milankovich Cycle is supported or enhanced by the natural emission of greenhouse gases. Most sensitive are the higher latitude regions, where, for a doubling of the amount of CO₂ in the atmosphere, we can have temperature increases in the order of 6 to 8°C, and that may lead to the thawing of the permafrost regions which I have already mentioned, which would create a positive feedback effect enhancing temperatures even more than just by temperature changes.

New studies indicate that the Arctic Ocean ice cover is about 40% thinner than 20 to 40 years ago, and there is dramatic climate change happening in the Arctic, about 2 to 3 times the pace for the whole globe and so this may lead to what I have already mentioned, to melting of the permafrost and another major positive feedback factor (Fig. 8, see p. 270).

Can we do something about it? If you say, reduce the emissions of greenhouse gases, and the way to go is get your energy from other sources, in the first place by energy savings, there is a lot that can be done there, renewable energy, nuclear energy, wind and solar power, and CO_2 sequestration is also a possibility. There is another possibility and that is to inject sulfur in the stratosphere, by bringing, for instance, H_2S with rockets and balloons in the stratosphere where you oxidise H_2S to SO_2 , which is further

oxidised to sulfuric acid, which forms sulfate particles that reflect sunlight. So this is, in principle, possible to do and I will show you the results of some calculations. I do this work in collaboration with scientists from the National Centre for Atmospheric Research, Philip Rasch and D.B. Coleman. The concept of doing this geoengineering goes back to Budyko and the study in the National Academy of Sciences in 1992, and more recent studies, the Teller proposal, then Govindaswamy and Caldera, and then I did a study which came out in August of this year. We use a General Circulation Model to estimate what might happen if you put some sulfur into the stratosphere in the form of sulfate particles that reflect solar radiation. We use a model with rather complete physics and simple chemistry but no biological feedback, so the permafrost story is not included in the model, also we hardly have a Carbon and Nitrogen Cycle. The circulation of the ocean is not changed because of our emissions of sulfur gases in the atmosphere.

Some further information about the model. The model goes up to 80 km altitude in the atmosphere, it is 52 layers, and has a special distribution of 2.5° by 2° latitude and longitude and about less than 1 km altitude speciation.

So now we do some experiments. We basically conduct four simulations (Fig. 9). In one simulation we work with the current atmosphere, fixed aerosol and greenhouse forcing, as happens at the moment in the atmosphere. Then, with the same initial conditions, we double the amount of CO_2 and then look at what happens to the average tempera-

Experimental Setup (part 3) Four Simulations performed Fixed aerosol and greenhouse forcing at present day values (Control) Doubled CO2 at beginning of simulation (2XCO2) Injection of 1 Tg S/yr as SO2 at 25km between 10N and 10S (Geo-sulfate) Doubled CO2 + Injection of SO2 (2XCO2 + Geo-Sulfate)

Figure 9.

ture; we already know the answer: it will go up. Then we do an experiment in which we inject 1 million Tg S/yr as SO₂ in the atmosphere at near 25 km altitude, in between 10°N and 10°S and that should lead to cooling, and I will show you that it does. And then we do both, we double the amount of CO₂ and we inject SO₂ and we look at the net result of that. Here (Fig. 10, see p. 271) we have the net result. The basic case, the control case is basically here, you can see it in yellow, but the model would predict average global temperatures of the order of a little above 288°K. If you double the amount of CO₂, but you do nothing with the sulfate particles, you see the rise in temperatures, a little over 2°C is the rise in temperatures in the stratosphere you go here, you have the cooling, also by about 2°C or more. If you do both, you end up with the black curve, you are almost back to the unperturbed conditions.

What I showed you before with temperatures, you can also see with precipitation (Fig. 11, see p. 271): doubling of CO_2 gives more rainfall, more particles in the atmosphere, less rainfall, all in mm/day and then you do both and you get basically the same initial state back. So, if the amount of CO_2 in the atmosphere doubles and temperatures go up, then we have the possibility, by adding sulfur to the stratosphere, to come back to normal temperature conditions.

The lifetime of the aerosol particles in the stratosphere is of the order of three to four years (Fig. 12, see p. 272). Normally it is shown that it is 1 to 2 years but, when you do the model calculation, it comes out to much longer. That means emissions into the stratosphere do not have to be as high as would otherwise be the case. The optical depth of the sulfate is about 0.06, which means that the sky will become a little lighter, but, on the other hand, you will also get wonderful sunsets and sunrises. It is basically a human volcano which is produced here. Precipitation changes around the globe are not very large, the average for the globe is 2.8, so the maximum deviation in this case would be 0.5, but these data are statistically not significant. On the whole, you can say that the precipitation changes have been less than 10% of the normal precipitation.

We can look at the temperatures (Fig. 13, see p. 273). If you do the doubling experiment of CO_2 you see the heating of the higher latitude regions of the atmosphere by the greenhouse effect, if you do doubling of CO_2 at the same time as sulfate in the atmosphere you get the white colour almost everywhere, meaning temperature changes between -1 and 1°C all over the globe, of the order of a few tenths of temperature changes in the atmosphere.

Let me stop here, we should definitively leave some time for discussion. This is, of course, an experiment which, at the moment, you only do on a computer. If we ever have to do a thing like that, it will only be if our climate runs away in some way. So you might think that it is very unlikely, but we have seen, in the case of the ozone hole story, that very unlikely things might happen, so we should be prepared by surprises in the future, which can only be discovered again by observations. The models are getting better. There is already considerable theoretical work going on concerning this sulfate experiment, more people are coming into action; until a few years ago or, basically, until this year this was a taboo thing, you should not study a thing like that, but this has changed now. There is a lot of activity taking place, you only do this when things are really getting very bad but we better be prepared that, if that is the case, in order to have some kind of weapon available to reduce the bad effects of other human activities.

Here I would like to stop: I am quite certain there will be very critical remarks but I thank you for your attention.

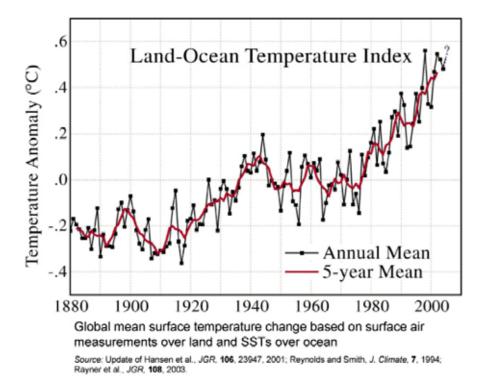
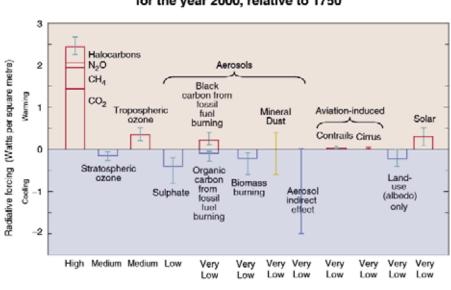


Figure 5.



The global mean radiative forcing of the climate system for the year 2000, relative to 1750

Level of Scientific Understanding



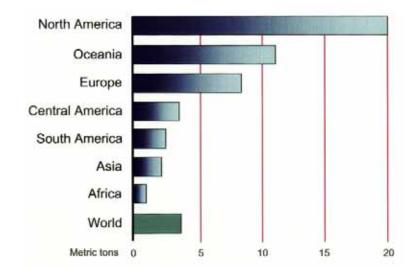
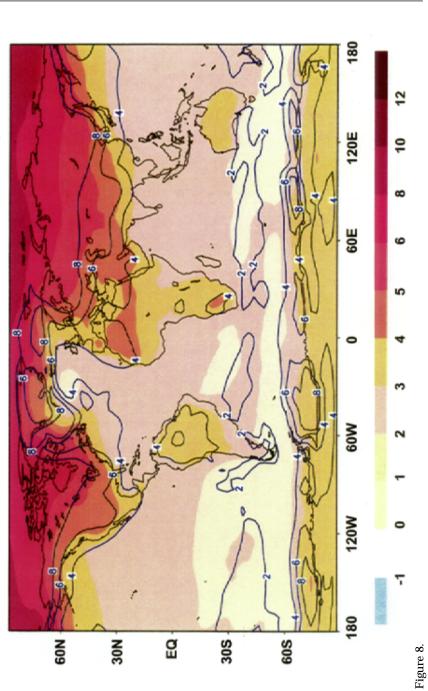


Figure 7.



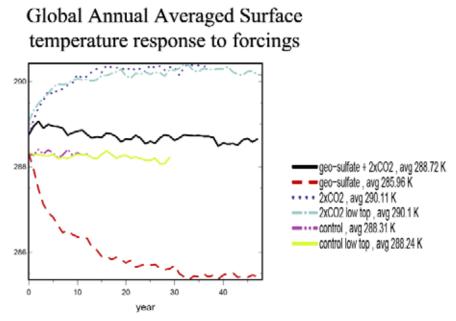
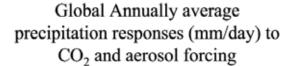


Figure 10.



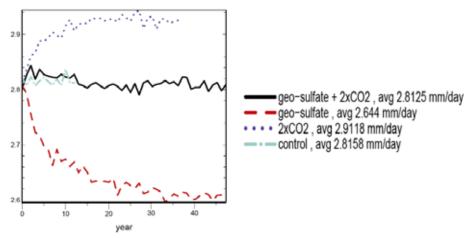
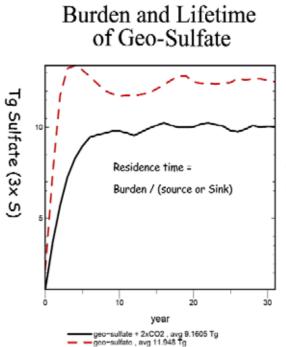


Figure 11.



Residence time ~ 3 years

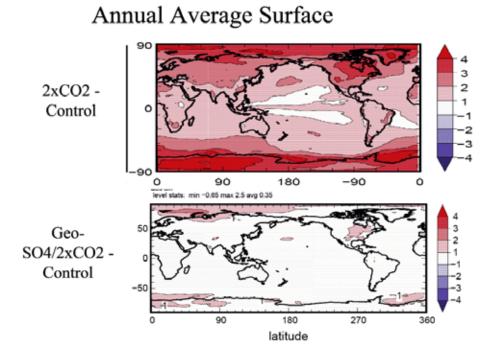
Residence time ~ 4 years

Residence time longer than usually estimated from volcanic aerosol (1-2 years)

Residence time depends on State! controlled by:

- cross tropopause exchange
- hydrologic cycle

Figure 12.



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Figure 13.