PHYSICS & LIFE

COLLANA DI FISICA APPLICATA ALLA BIOLOGIA
ALLA MEDICINA, ALL'AMBIENTE, AI BENI CULTURALI
ALLE ATTIVITÀ MOTORIE E SPORTIVE E ALLE SCIENZE GASTRONOMICHE

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La ricerca della verità è più preziosa del suo possesso

Albert Einstein

Physics in every day life è uno dei libri fondanti della Fisica applicata alla Biologia, alla Medicina, all'Ambiente e, successivamente, ai Beni culturali, e da poco alle Attività motorie e sportive ed alle Scienze gastronomiche. In altre parole a tutti quei campi in cui quotidianamente ci si confronta inconsciamente con quel che avviene intorno o con quanto ci si trova a fare nella vita per lavoro, per passione o per relax, solitamente senza rendersi conto di come invece, sempre e comunque, la Fisica svolge un ruolo ben preciso ed ha multiformi riscontri. La conoscenza dei quali può rappresentare un importante strumento per meglio operare, od addirittura uno stimolo per indirizzare od approfondire la propria ricerca. Obiettivi questi che la collana si propone di affrontare e stimolare.

Web content

Daniele Visioni

A climate engineering technique for a warming planet

Stratospheric sulfur injection as a temporary solution to greenhouse gasses increase

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Chapter 1

Introduction

1.1 Climate change and our planet's future

Anthropogenic global warming (AGW) is one of the main problems that humanity is facing in the present, and its preponderance is likely to grow even more in the course of the 21^{st} century [1]. But what do we mean by "Global warming"? And why do we define it as "Anthropogenic"?

The first answer is that in the last century we have observed an increase in the surface temperatures of our planet, particularly during the last 50 years. This increase has been detected in many ways, through direct surface measurements and satellites observations. The time span of this warming, together with the -always increasing- precision of the measurements, has allowed the scientific community to understand that such warming is not due to normal oscillations of the climatic system [2]. Furthermore, the warming is consistent with climate simulations across a large number of climate models [3]. This warming has caused a sharp reduction in sea ice extent in the Arctic (itself connected with an increase in the global mean sea level) and an increase in the frequency of extreme weather events [1].

The causes of this warming can be traced back to the increase in greenhouse gases in the planet atmosphere: in the last two centuries, atmospheric CO_2 has steadily increased [4], and at a rate that has never been observed in all the period where humans lived on this planet, as shown by ice core sample measurements [5] that allow us to go back thousand of years to explore past atmospheric composition. This increase is not limited to just CO_2, but extends to all greenhouse gases [6]. Greenhouse gases (GHGs) are not inherently dangerous. Indeed, if they were not present at all in our atmosphere, our planet would be much cooler (around 260 K), even if they represent a very small portion of all the constituents of the atmosphere (less than 0.04 %) The modern increase, measured for CO_2 from 280 ppm in the 19^{th} century to 380 ppm in the year 2000, has thus changed the Earth radiative balance greatly. But why has it increased so much? The answer is that this increase is anthropogenic in nature, that is, the human footprint is clear. The amount of carbon in the Earth

atmosphere is regulated by many factors, with the most important two being: 1) the biogenic carbon cycle, (on land and in the oceans) where plants or algae constantly remove carbon from the atmosphere through photosynthesis and emit carbon into the atmosphere through natural processes, including respiration and decay and 2) the solubility pump, where carbon is dissolved in the ocean by CO_2 reaction with water. A large part of the carbon present on the planet is however normally stored below the planet surface as coal, oil and natural gas, formed through fossilisation and decomposition of organic matter over millions of years.

When, during the Industrial revolution, humanity started burning these fossil fuels, the carbon stored there was released in the atmosphere, and at a (growing) rate that plants and oceans have been unable to capture. Once CO_2 is released in the atmosphere, furthermore, it is virtually indestructible, having an atmospheric lifetime of more than a century, since it reacts little with other atmospheric components. For this reason, even an immediate stop to fossil fuel burning would not have immediate effect on the amount of CO_2, and any proposed mitigation scheme for human emissions would take century before yielding results. The strong correlation between anthropogenic activities and the amount of CO_2 in the atmosphere can be precisely tracked measuring the amount of ^{14}C, thus detecting how much carbon comes from the biogenic cycle and how much from fossil fuels [7] and making any attempt to deny the influence of humanity on the increase of carbon dioxide scientifically very difficult. Given this notion, the next question could be: what about the future?

Clearly, our society has no intention to give up fossil fuels in the blink of an eye. To make matters worse, human population is constantly increasing, and so are CO_2 emissions from developing countries that are improving their standard of living. So large changes are still possible, and different futures can be imagined, from more pessimistic ones (emissions keep building up) to more optimistic ones (we manage to reduce and finally stop new emissions in the next century). To translate this into a quantifiable prediction that can be used also in climate models, the Intergovernmental Panel on Climate Change (IPCC) has developed, bringing together scientists from different fields, what are known as Representative Concentration Pathways (RCP) [8]. These scenarios' main difference is in the different overall radiative forcing (RF) produced by the GHGs in 2100: namely, the scenarios range from RCP2.6 (GHGs forcing peaking at 3 W/m^2 and decreasing to 2.6 W/m^2 by the end of the century) to RCP8.6 (GHGs forcing at 8.5 W/m^2 by 2100, the scenario that is expected if emissions continue rising as they are now, known as "business as usual"). In Fig. 1.1, a schematic representation of the three scenarios is shown.

As was said before, RCP scenarios merely prescribe for the future different emissions scenarios. It is the job of climate modellers to integrate these prescriptions into climate models, in order to understand the changes that the increases in RF might bring. A compendium of the possible impact of the different emission scenarios, from surface temperature increase to its effects, spanning from sea level changes to desertification and increase of sever events

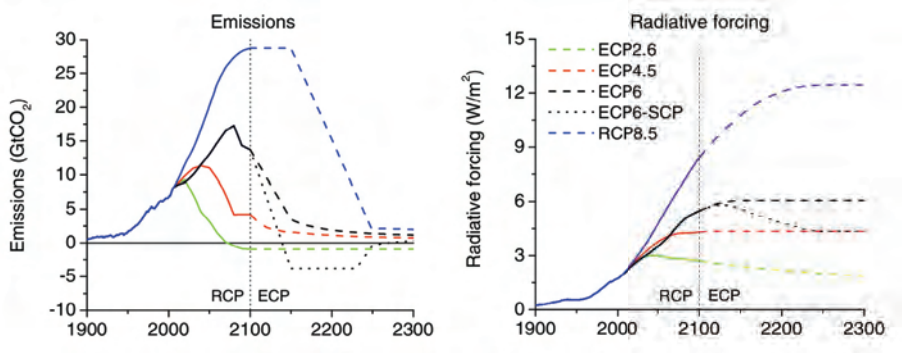

Figure 1.1: Emissions of CO_2 (left, in $GtCO_2$) and related RF (right, in W/m^2) for the three scenarios plus a transition scenario between RCP6.0 and RCP4.5. All scenarios are extended up to 2300 (Extended Pathway Scenarios). Figure taken from [8].

in various areas, can be found in [9]. In light of these scenarios, it is easier to understand some of the international agreements that have been reached in the last years, such as, for instance, the Paris Agreement signed in 2016 (`https://unfccc.int/sites/default/files/english_paris_agreement.pdf` the link to the official signed agreement). In this agreement, most of the nations agreed over:

> "Holding the increase in the global average temperature to well below 2°C above pre-industrial levels and pursuing efforts to limit the temperature increase to 1.5°C above pre-industrial levels, recognizing that this would significantly reduce the risks and impacts of climate change [...]"
>
> (Paris Agreement, Art. 2.1.a)

As can be clearly seen from Fig. 1.2, such an ambitious goal could only be achieved under the most optimistic of all the emission scenarios. How likely this is to happen is an interesting political discussion, however it can be agreed that it is going to be an hard objective to achieve, considering the drastic curb in emissions (and thus, a drastic change in the way we define civilization) that it requires. So at this point, the discussion leads us to the last question which introduces the main objective of this work.

Is there anything else humanity (in general) and the scientific community (in particular) can do to achieve the goal of keeping the global men warming below 2 °C? In other words, aside from hoping for a new technological breakthrough that could completely free us from the dependency on fossil fuels, is there a chance to somehow manage the temperature of the planet in a way that our survival on this planet is assured? Any possible proposed solution that is considered an answer to this question, and as we will see there are many, can be summed up under the term "geoengineering". With this term we define any human effort

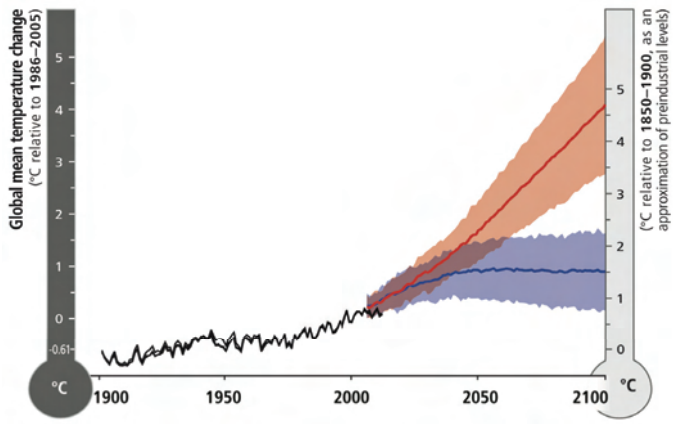

Figure 1.2: A comparison of surface temperature changes from 1986 to 2005 (black line) and the predicted temperature changes under the most pessimistic emission scenario (RCP8.5, red line) and most optimistic emission scenario (RCP2.6, blue line). Figure taken from [9].

(more or less technological: from space mirrors to planting more trees) aimed at managing the planet climate in order to keep it hospitable for human life. As of now, and for the foreseeable future, very few of these proposals have ever been tested even on a small scale (such as some forms of carbon dioxide removal), but most of them remain purely hypothetical.

In the next section, we will give an overview of some of the most important geoengineering methods that have been proposed by the scientific community in the last years.

1.2 Geoengineering techniques: an overview

Climate engineering, more commonly known as geoengineering, has been defined as "as the deliberate large-scale intervention in the Earth's climate system, in order to moderate global warming." [10]. Geoengineering is not, in itself, a new idea. While weather modifications date back to at least the 19^{th} century, particularly in the United States, real geoengineering proposals date at least to 1965 with a preliminary report from the US President's Science Advisory Council was issued. An in-depth review of the history of geoengineering can be found in Keith (2000) [11].

Most methods can be divided in two different categories: Carbon Dioxide Removal (CDR) methods and Solar Radiation Management (SRM) methods. In the first category are all the proposed technological means of directly removing CO_2 or other GHGs from the atmosphere. In the second category are all methods that aim to reduce the amount of solar radiation arriving at, or

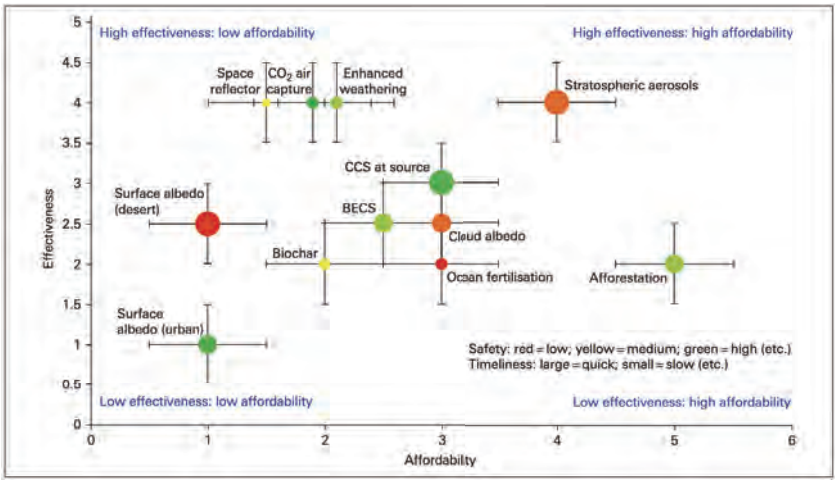

Figure 1.3: An evaluation of the most well known geoengineering methods over four different parameters: Effectiveness, Affordability, Safety and Timeliness. Figure taken from [10].

being absorbed by, the surface of the planet. Geoengineering methods can be classified, as shown in Fig. 1.3, in terms of four parameters:

1. Effectiveness: how efficient is the proposed method to actually cool the planet.

2. Affordability: how costly is the method.

3. Timeliness: how fast can the method be implemented and, more in general, how long would it take to develop the actual functioning method.

4. Safety: how safe is the method, in terms of possible global modifications of the environment, reversibility of the method and impact on the population.

As can be seen from Fig. 1.3, that lists 12 methods overall, there is a lot of variation between them. There are costly methods with very little effect (such as large scale albedo modifications, that is painting cities and roads white in order to reflect a larger part of the incoming solar radiation); simple methods such as afforestation, that is the increase in large vegetation that can act as sink of CO_2 and very effective methods such as building huge space reflectors to be put in orbit around the planet that sound more like something one would find in a science-fiction book. Lastly, the method that will be discussed in this thesis stands a bit isolated from the rest, in the uppermost right corner. The method, labeled as 'Stratospheric aerosol', aims to reduce part of the incoming solar radiation by injecting aerosol (in our case it will be sulfate aerosols, but

others have been proposed) in the stratosphere, in order to act as a reflector of the sunlight. The method is rated as rather affordable and effective, quick but a low safety. The aim of this work will be centered around this problem, that is, understanding and studying some of the possible side effects of sulfate geoengineering (SG).

Compared to other geoengineering methods, one of the positive points of SG is the existence of natural experiments that have produced, in the past, similar effects to the one that is desired. Indeed, when the idea was first proposed, first by Budyko in 1978 [12] and then by Crutzen in 2006 [13], their reasoning started from a simple observation: when tropical large volcanic eruptions have happened, the sulfate aerosols that were injected in the stratosphere produced an optically thick cloud that persisted for months, producing a cooling of the surface temperatures that was detected even one year after from the eruption. Furthermore, even if the injection of sulfate caused by a volcanic eruption increases the aerosol load in the stratosphere by one or two orders of magnitude (for the last large volcanic eruption, Pinatubo in 1991, it is estimated to have been 7 or more Tg of sulfur), this amount is only a small percentage of the entire sulfur budget in the entire atmosphere (estimated to be around 90 Tg-S/yr [14], [15]).

With this in mind, it is easy to see why the idea of artificially injecting sulfate in the stratosphere in order to cool the planet seems, at first glance, a good idea. It is a compound already present in large quantities, and we have a natural proxy to study in order to evaluate some of the possible effects. However, before moving from an idea to its realization, expecially for such a global endeavour, an in-depth study is necessary to understand what would happen if ever it was actually applied in the real world. In order to do this, exactly has it is done for the different RCP scenarios, climate models are the best way in which eventual shortcomings, difficulties and possibly harmful side-effects can be highlighted before eventual testing in the real world. The goal of this book is to show the results that we have obtained regarding the investigation of some of the possible side effects of SG on the atmosphere. To do so, I (with the support and guidance of the principal developer of the model and my advisor. Prof. G. Pitari) have performed and analysed simulations with the University of L'Aquila Climate-Chemistry Model (ULAQ-CCM). The following chapters are structured in this way: in Chapter 3, I illustrate some of the results obtained with ULAQ-CCM regarding past explosive volcanic eruptions, analysing the most important climatic effects and comparing our results with available measurements.

In Chapter 4 I show the results we obtained under a future SG scenario, focusing on the main aspects that could produce a significant RF change, thus affecting the intended surface cooling that would be needed to offset the warming produced by GHGs. In particular, we will show the changes that stratospheric sulfate would produce on key atmospheric chemical components (ozone, methane and other GHGs) both because of chemical and transport changes, interactions of the injected sulfate with dynamical processes in the

stratosphere and modification of upper troposphere cirrus cloudiness. In the Conclusions we give an overview of the state of the art regarding sulfate geoengineering research, beyond the studies performed with our model.

All the results shown in Chapter 4 have already been published (with the author of this work as first author) in four articles in the peer-reviewed journal Atmospheric Chemistry And Physics, as part of the special issue "*The Geoengineering Model Intercomparison Project (GeoMIP): Simulations of solar radiation reduction methods*" (the entire special issue can be found here `https://www.atmos-chem-phys.net/special_issue376.html`).

Lastly, in Appendix A, we offer an in-depth description of the ULAQ-CCM composition-climate model with particular focus on the most important components used for assessing the effect of a stratospheric sulfur injection, together with a validation of some of its modelling skills done in the framework of some international multi-model evaluations (Chemistry-Climate Model Validation, CCMVal2 and Chemistry-Climate Model Intercomparison, CCMI).

Chapter 2

Volcanic eruptions A natural experiment

Explosive volcanic eruptions are not such a rare occurrence on our planet. In the last 250 years, 13 eruptions with a Volcanic Explosive Index (VEI) larger than 4 were counted [17], with the last one (Pinatubo) happening in 1991. Indeed, the last 27 years can be considered a rather long quiescent period compared to the usual frequency of those kind of eruption. For this reason, opportunities to study the impact of explosive volcanic eruptions are limited to those available after the Pinatubo eruption, with all the limitations of measurements (both in-situ and from satellites) taken 27 years ago, and, as a consequence, an in-depth understanding can be gained only by relying heavily on climate models simulations. Climate records ([18]) have demonstrated that major volcanic eruptions reaching the stratosphere may globally cool the surface by a few tenths of a degree. The stratospheric aerosol layer may also be perturbed by weaker eruptions ($\sim$0.5-3 Tg-SO_2 injected in the upper troposphere and lower stratosphere), with small but non-negligible impact on global surface temperatures ([19]). These eruptions have contributed to the generally increasing levels of stratospheric aerosols between 2002 and 2010 ([20]; [21]), as from measurements by lidars and sun-photometers.

Large explosive volcanic eruptions may significantly affect the planetary scale transport of stratospheric trace species, by considerable amounts of particles and sulfur gases injected above the tropopause. Perturbations of stratospheric dynamics may result from both local stratospheric heating (with associated changes of temperature and zonal wind fields), as well as from climate changes following the increasing scattering of incoming solar radiation by the volcanic aerosols ([22]; [23]). The radiatively forced changes of the stratospheric circulation during the first two years after the eruption of Mt. Pinatubo (June 1991) may help explain the observed trend decline of long-lived greenhouse gases (as CH_4 and N_2O) ([24]; [25]), as a result of the increased mid- to high-latitude stratospheric downward fluxes. Since the stratosphere contains lower CH_4 and N_2O mixing ratios, a decline in the observed trends could result from a higher degree of exchange between the stratosphere and the troposphere ([26]).The

initial SO_2 plume located in the lower stratosphere ([27]) undergoes rapid OH oxidation, thus increasing the amount of gas phase H_2SO_4, followed by its nucleation into H_2O-H_2SO_4 ultrafine particles. These end up in accumulation mode aerosols after self-coagulation and H_2SO_4 condensation (i.e., gas-particle conversion). An optically thick cloud of supercooled sulfate aerosols is finally produced ([28]; [29]), which is highly reflective in the visible and UV. This effect is particularly important in the tropics, where the aerosols (depending on the quasi-biennial oscillation phase) may remain confined for several months after the eruption with relatively high values of optical thickness. A typical example of a tropical eruption with a rather pronounced particle confinement in the tropical lower stratosphere is the 1991 Pinatubo case ([30]). The stratospheric radiative impact of volcanic aerosols produces additional diabatic heating rates through the following mechanisms: (a) direct interaction with solar near-infrared and planetary far-infrared radiation and indirect ozone absorption of the aerosol diffused UV-visible solar radiation ([31]; [32]); (b) changing ozone absorption of incoming solar and outgoing planetary radiation, as a consequence of ozone losses produced by heterogeneous chemistry on sulfuric acid aerosols [22] ([33]) and perturbed photolysis rates and stratospheric large scale transport ([34]). A 2-3 K warming in the tropical lower stratosphere was measured in September-October 1991 after the Pinatubo eruption ([35]) and a 20-25% ozone depletion in the 16-28 km layer over the tropics during October-November 1991 ([36]). A model study by [37] calculated a global ozone column decrease by about 7 DU because of the Mt. Pinatubo eruption. Climate changes are due to increasing scattering of the incoming solar radiation by the volcanic particles, which results in a surface cooling ([38]). The 1991-1992 time series of the calculated globally averaged net radiative flux change at the tropopause due to Pinatubo aerosols shows that the largest change is occurring during January 1992 (-4.5 W/m^2, for clear sky conditions and 0.15 optical thickness at λ=0.55 μm) and decreasing afterwards with an e-folding time of approximately 12 months ([22]). Upper stratospheric dynamical anomalies related to major volcanic eruptions have also been detected and highlighted. A study of [39] on the stratospheric dynamical perturbations during 1982/83 (after the El Chichón eruption) shows how anomalies of temperature and zonal winds in the upper stratosphere could be explained as a consequence of the adiabatic cooling taking place in the tropical stratosphere due to the increased upwelling. A decrease of temperature was found at an altitude of about 40 km, although a slightly positive diabatic anomaly was present there. Changes in diabatic heating rates due to volcanic aerosols have the direct effects of warming up the tropical lower stratosphere and upsetting the stratospheric dynamics, because of an increased extra-tropical planetary wave activity associated to the lower stratospheric anomalies of the mean zonal wind field. An additional residual upwelling motion is produced in the tropical stratosphere of the order of 10-20% of the unperturbed vertical velocity at approximately 30 hPa ([31]).

Several modelling studies have been conducted to investigate the effects of volcanic aerosols on climate and on the stratospheric circulation, looking also

at the feedback of the aerosol induced dynamical changes on the volcanic cloud dispersal ([22]; [40]). Examples of more recent modelling re-analyses of the Pinatubo perturbation are also available in the literature ([41]; [42]; [43]). As discussed so far, the planetary scale transport of stratospheric species and the dispersal of volcanic aerosol clouds may be significantly affected by the radiative perturbations induced by the volcanic aerosols themselves. The phase of the quasi-biennial oscillation (QBO) of equatorial mean zonal winds, however, is even more important for stratospheric trace species transport, including volcanic aerosols transport. [30] have studied the tropical stratospheric circulation using satellite aerosol data and have clearly shown the differences between dynamical regimes during dominant easterly or westerly shear of the QBO. The streamlines of the QBO circulation show that definite upwelling motion is extended everywhere in the tropical stratosphere (from the tropopause up to 30 km altitude) during years with dominant easterly shear. On the other hand, during years with dominant westerly shear, descent relative to the mean stratospheric circulation occurs over the equator and more aerosols can be transported down to altitudes within a few kilometres above the tropopause, where poleward isentropic transport is relatively fast. The main conclusion of [30] is that when the QBO easterly shear is present (as in the months following the Pinatubo eruption), aerosols are lofted over the equator and less transport towards the poles is obtained with respect to the case of westerly shear. The additional upwelling due to the Pinatubo aerosol radiative heating has therefore a feedback on the dynamics of the middle atmosphere similar to that of the QBO easterly shear and may additionally help to confine the particles in the tropical reservoir and to increase their lifetime.The interconnection between high loadings of tropical lower stratospheric aerosols and the QBO is not only visible in the different features of the cloud dispersal towards mid-high latitudes and the mid stratosphere, (depending on the phase of the oscillation), but also in the possibility that a sustained high loading of sulfate aerosols may modify the natural evolution of the QBO. This could be the case of planned experiments of sulfate geoengineering by means of SO2 injection in the equatorial lower stratosphere of the order of 5-10 Tg-SO_2/yr [40] ([44]). In their study [44] found that such an injection dramatically alters the quasi-biennial oscillation, prolonging the phase of easterly shear with respect to the background atmosphere without geoengineering sulfur injection. This is caused by the increased aerosol heating, and associated warming in the tropical lower stratosphere and higher residual vertical velocity. In the case of maximum perturbation, the lower tropical stratosphere could even result to be locked into a permanent westerly QBO phase.

In the following chapter, we focus on studying the volcanic aerosols effect on the atmosphere using the ULAQ-CCM model. For an in depth description of the model we remand to appendix A. For sections 2.1 and 2.2 we will focus in particular on the effect of explosive volcanic eruptions. In section 2.3, on the other hand, we focus on sulfate originating from non-explosive volcanoes, analyzing the chemical-radiative effects of this constant source of sulfate.

2.1 Stratospheric sulfate interaction with the atmosphere

2.1.1 Numerical experiment set-up

The model has been run for a total of 20 time-dependent simulations from 1960 to 2000. Of these numerical experiments, 10 will be now on referred to as 'reference' simulations (REF): they do not include the volcanic aerosols, except in the heterogeneous chemistry code, through use of externally specified time-dependent fields of sulfate aerosol surface area density (SAD) ([45]). The other 10 experiments will be referred to as 'volcanic perturbed' simulations (VE) with inclusions of the volcanic aerosols both in heterogeneous chemistry and in the radiative transfer code. Table 2.1 shows a summary of the major explosive eruptions from 1960-2000 included in the present study; the four tropical cases will be analyzed in detail.

Table 2.1: Major volcanic eruptions considered here, with the adopted amounts of the injected SO_2 and indication of the QBO E/W shear at the eruption time and following months. The vertical layer of SO_2 injection is assumed between 90 and 40 hPa (∼16-22 km altitude), with a peak at 60 hPa (∼18 km altitude).

Eruption	**Time**	**Tg-SO_2**	**QBO E/W shear**
Agung (8°S, 11 °E)	16 May 1963	12	W
El Chichón (17°N, 93 °W)	4 April 1982	7	W
Nevado del Ruiz (5°N, 75 °W)	13 November 1985	1.2	E
Pinatubo (15°N, 120 °E)	16 June 1991	20	E

Figure 2.1 shows the calculated time series of the stratospheric aerosol optical depth (AOD) at λ=0.55 μm, compared with reference values available at NASA/GISS (`http://data.giss.nasa.gov/modelforce/strataer`). The three largest eruptions (Agung 1963, El Chichón 1982, Pinatubo 1991) produce maximum stratospheric AOD ranging between 0.06 and 0.10 in the ULAQ-CCM, which are somewhat underestimated with respect to the NASA/GISS values. Note that the eruptions of Fernandina (1968) and Tolbachik (1975) have not been included in the model simulation. The calculated AOD is obtained by vertically integrating the aerosol extinction. This is in turn obtained from the aerosol size distribution calculated in the ULAQ-CCM aerosol microphysics module, with final conversion of the particle concentration into aerosol extinction by means of a Mie scattering program ([47]).