Doctoral Thesis

Cloud and Surface Responses to Stratospheric Aerosols following Major Volcanic Eruptions

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Cloud and Surface Responses to Stratospheric Aerosols following Major Volcanic Eruptions

A dissertation submitted to the
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presented by
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Abstract

Major volcanic eruptions may inject large amounts of gases and aerosol particles into the lower stratosphere. Within weeks after an eruption, layers of aqueous sulfuric acid droplets will form from the sulfur dioxide precursor gas contained in the plume. The aerosol reduces the transmission of solar radiation towards the troposphere and can thereby significantly affect air temperature, evaporation and precipitation at the Earth’s surface. At the same time, the influx of stratospheric aerosol droplets, which are typically larger than the upper tropospheric aerosol particles, into the comparatively moist upper troposphere may trigger the homogeneous freezing of the droplets and subsequent ice particle growth. It has been hypothesized, therefore, that the stratospheric sulfate aerosol may affect the abundance and the optical properties of ice clouds when it reaches the upper troposphere.

With respect to these two volcanic influences, the objectives of this thesis were (1) to assess how well state-of-the-art general circulation models (GCMs) capture surface climate anomalies observed after strong volcanic eruptions, and (2) to analyze backscatter measurements taken by the satellite-borne CALIOP lidar — one of the most suitable instruments currently available for global aerosol-cloud studies — for evidence of a modification of the optical properties and abundance of ice clouds by sulfate aerosol droplets of stratospheric origin.

In Chapter 2 of this thesis, I compare 17 GCMs with each other and with observational data following two major volcanic eruptions. After statistically removing the El Niño/ La Niña-Southern Oscillation (ENSO) surface climate impact, I found that the 17 investigated models successfully simulate the observed 4-months delay in the surface air temperature responses to the ENSO phase, but simulate somewhat too fast precipitation responses during the El Niño onset stage. The observed correlation between surface air temperature and ENSO phase (correlation coefficient: 0.75) is generally captured well by the models (simulated correlation coefficients of 0.71, ensemble means of 0.61 – 0.83). With regard to the precipitation over tropical land regions, the mean correlation coef-
ficients with the ENSO phase are $-0.59$ (observations) and $-0.53$ (models). However, individual ensemble members have correlation coefficients as low as $-0.26$.

I find that the observed ENSO-removed tropical land temperature and precipitation decreased by about 0.35 K and 0.25 mm/day after the 1991 Pinatubo eruption, whereas no significant decrease in either variable was observed after the El Chichón eruption in 1982. The investigated models generally capture this behavior, despite a tendency to overestimate the precipitation response to El Chichón. There is substantial scatter both across the models and across the ensemble members of individual models, which implies that natural variability may still play a prominent role, despite the strong volcanic forcing.

In Chapter 3 of this thesis, I analyze eight years of CALIOP lidar backscatter measurements, taken from ice clouds between $82^\circ$ N and $82^\circ$ S, searching for signs of a sulfate-aerosol effect on the backscatter distributions, occurrence frequencies and altitudes of cirrus clouds and the glaciated parts of mixed-phase clouds. To this end I have focussed on the eruption of the Nabro volcano in Northeast Africa in June 2011. This event released 1–1.5 Mt of sulfur dioxide into the lower stratosphere, making it the largest volcanic eruption since Pinatubo in 1991 in that respect. I found that the stratospheric aerosol that formed after the Nabro eruption did not modify the optical properties, abundance or residence altitudes of ice clouds in a statistically significant manner on a global scale, not even in the uppermost troposphere. Accordingly, the Nabro eruption induced no detectable volcanic aerosol-ice cloud radiative forcing effect in the four seasons following it.

Simulations of the Nabro sulfate aerosol plume performed with the global chemistry-climate model SOCOL-AER indicated number density increases of the largest aerosol droplets by more than a factor of 3 at the lower edge of the plume near the tropopause in the months after the eruption. As the largest droplets of any sulfate aerosol population are the ones expected to freeze first, our results suggest that the optical properties and the abundance of ice clouds are influenced at most weakly by volcanic eruptions. Our findings are consistent with previous studies which suggested that temperatures and cooling rates are the main determinants of the ice particle number density in ice clouds, whereas number densities and sizes of sulfate aerosol droplets play only a secondary role.

Finally, in Chapter 4 I used the CALIOP dataset to validate the occurrence frequencies of ice clouds in the aerosol-cloud GCM ECHAM6-HAM2, which is presently one of the most advanced aerosol-cloud GCMs. I found that there is generally a good match between the simulated and the observed ice-cloud occurrences. However, the simula-
tions show that ice-cloud abundance is substantially underestimated in the tropics and
overestimated in regions south and west of the Taklamakan desert as well as over parts
of Antarctica. Moreover, the altitudes of the highest ice-cloud tops are overestimated
by 1–2 km at most latitudes, which may bias the ice-cloud radiative forcing by several
W/m².
Zusammenfassung


Die vorliegende Arbeit hatte in Bezug auf diese zwei vulkanischen Einflüsse das Ziel, (1) zu bewerten, wie gut aktuelle globale Zirkulationsmodelle nach starken Vulkanausbrüchen an der Erdoberfläche beobachtete Klimaanomalien simulieren, und (2) von dem satellitenbasierten Lidar CALIOP gemachte Rückstreumessungen auf Anzeichen für eine Modifikation der optischen Eigenschaften und der Häufigkeit von Eiswolken durch stratosphärische Sulfataerosoltröpfchen hin zu untersuchen. CALIOP ist derzeit eines der geeignetsten Instrumente für globale Aerosol-Wolken-Studien.

In Kapitel 2 dieser Arbeit verglich ich 17 globale Zirkulationsmodelle miteinander und mit Beobachtungsdaten nach zwei starken Vulkanausbrüchen. Dabei habe ich nach Entfernen des klimatischen Einflusses der El Niño-Southern Oscillation (ENSO) herausgearbeitet, dass die 17 untersuchten Modelle die beobachteten viermonatigen Verzögerungen der 2-Meter-Lufttemperatur, mit der diese auf die ENSO-Phase reagiert, erfolgreich wiedergeben, aber andererseits eine etwas zu schnelle Reaktion des Niederschlags in der Anfangsphase von El Niños simulieren. Die Modelle simulieren die beobachtete Korrelation zwischen der 2-Meter-Lufttemperatur und der ENSO-Phase (Korrelationskoeffizient von 0.75) insgesamt erfolgreich (simulierte Korrelationskoeffizienten
von 0.71, Ensemble-Mittelwerte: 0.61 – 0.83). Die mittleren Korrelationskoeffizienten zwischen dem Niederschlag und der ENSO-Phase sind −0.59 (Beobachtungen) und −0.53 (Modelle). Einige Ensemble-Elemente weisen jedoch sehr geringe Korrelationen auf (Korrelationskoeffizienten bis −0.26).


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Chapter 1

Introduction

First measurements of stratospheric aerosol properties were made by Junge et al. (1961) who discovered the stratospheric aerosol layer using a balloon-borne particle impactor. Junge found that stratospheric aerosols are mainly made up of aqueous sulfuric acid droplets. In the lower stratosphere, these particles have mean diameters of about $150 \text{ nm}$ and number densities of about $10 \text{ cm}^3$ in volcanically quiescent time periods, and about 2–4 times larger diameters and temporarily much larger number densities following large volcanic eruptions (Deshler et al., 2008).

Stratospheric aerosol droplets form after and while sulfur-containing precursor gases and particles of either volcanic or anthropogenic origin reach the lower stratosphere. Formation of new particles, termed gas-to-particle conversion, is of particular importance in the tropical tropopause region. The most important precursor gases are sulfur dioxide (SO$_2$) and carbonyl sulfide (OCS) (SPARC, 2006; Sheng et al., 2015). Following a large volcanic eruption, freshly nucleated sulfate droplets form on time scales of weeks to months and evolve in size by condensation and evaporation in the lower stratosphere (SPARC, 2006). They are transported east- and westward by zonal stratospheric winds, and polewards by the Brewer-Dobson circulation. Their removal from the stratosphere occurs by exchange of air through tropopause folds and by sedimentation, as illustrated in Figure 1.1. Though humans may influence the stratospheric aerosol layer by increased surface emissions of SO$_2$ as measured over parts of mainland Asia over the last decade, no long-term trend in the non-volcanic stratospheric aerosol has been observed so far (SPARC, 2006; Deshler et al., 2008).

The optical depth $\tau(\lambda, z)$ is a dimensionless quantity that can be used to quantify the stratospheric aerosol burden. It measures the attenuation of light at wavelength $\lambda$ travelling a distance $z$ through a medium, like an aerosol layer. Figure 3.2 of Chapter 3 shows a Hovmöller diagram of the stratospheric aerosol optical depth (AOD) of the
Chapter 1 Introduction

Figure 1.1: The life cycle of the stratospheric aerosol, from SPARC (2006).

years 1990–2011. Clearly visible is the AOD increase following the eruption of Mount Pinatubo in June 1991 at 16°N. Figure 1.2 is a photography taken two months after the eruption from a NASA space shuttle. It shows two distinct aerosol layers in the lower stratosphere that strongly affect the extinction of visible sunlight. The AOD can be defined by Beer-Lambert’s law,

\[ I(\lambda, z) = I_0(\lambda)\exp(-\tau(\lambda, z)) \]  \hspace{1cm} (1.1)

where \( I_0(\lambda) \) is the intensity of the light incident on the aerosol layer and \( I(\lambda, z) \) is the intensity of the light that is transmitted at a geometrical depth \( z \) into the layer. The optical depth at wavelength \( \lambda \) can be defined as

\[ \tau(\lambda, z) = \int_0^z \varepsilon(\lambda, z') \, dz' \]  \hspace{1cm} (1.2)

where \( \varepsilon(\lambda, z) \) is the extinction at that wavelength. The extinction \( \varepsilon \) of the stratospheric aerosol layer can be determined from, from example, measurements by the Stratospheric Aerosol and Gas Experiment (SAGE) II instrument, though substantial uncertainties may be present for the months immediately following a large eruption (Arfeuille et al., 2013).

**Climate impacts of large volcanic eruptions.** After major eruptions, more outgoing and solar infrared radiation is absorbed in the stratospheric aerosol layer than in volcanically quiescent periods, while at the same time more solar radiation is scattered
back to space. The resulting net effect is a warming of the stratospheric aerosol layer and the air masses adjacent to it, as indicated in Figure 1.3. Such heating, as observed after the Pinatubo eruption, can affect the dynamics of the lower stratosphere (McCormick et al., 1995). Increases in stratospheric aerosol burden following strong eruptions also provide larger surface area densities for heterogeneous chemistry reactions to occur, which may result in ozone loss (Heckendorn et al., 2009). Moreover, as an optically thicker stratospheric aerosol layer scatters a larger fraction of the incoming solar radiation back to space, this may result in tropospheric cooling and decreases of global surface air temperatures. A global lower tropospheric cooling of about 0.5 K was observed after the Pinatubo eruption (Dutton and Christy, 1992). Climate model simulations of Robock and Liu (1994); Broccoli et al. (2003) indicated significant precipitation decreases in the tropics and on a global scale, caused by reduced evaporation, in the year following a major eruption. Gu and Adler (2011); Trenberth and Dai (2007) analyzed several decades of satellite-derived precipitation data and found substantial reductions in global land precipitation following the eruptions of El Chichón and of Pinatubo. Major eruptions are thought to affect the monsoon circulation. Modelling studies indicate decreases in the Asian monsoon precipitation following eruptions in the NH high latitudes Oman et al. (2005); Schneider et al. (2009). Further observed and simulated surface climate responses to large volcanic eruptions were reviewed by Robock (2000); Timmreck (2012); Robock (2013); Langmann (2014). Such climatic effects may last from months to a few years, owing to the small size and the resulting small sedimentation speed of the aqueous
sulfuric acid droplets. Volcanic ash particles, on the other hand, are thought to have negligible climatic effects because the majority of them have large sizes and correspondingly short stratospheric residence times (Robock, 2000).

Chapter 2 examines surface air temperature and precipitation responses to two recent large eruptions, El Chichón in April 1982 and Mount Pinatubo in June 1991, as simulated by 17 state-of-the-art climate models as part of the Coupled Model Intercomparison Project Phase 5 (CMIP5). The chapter places emphasis on the separation of the climate responses to the eruptions from the responses to concurrent El Niño events.

**Scattering of light by small particles.** The extinction of light in the atmosphere is the result of scattering and absorption by gas molecules (mostly N₂ and O₂) and particles suspended among them, in particular cloud droplets, ice crystals and aerosol particles such as mineral dust grains. All these molecules and particulate matter can be considered as dielectric material, meaning that they respond to incoming light by forming dipoles and thus by generating an electromagnetic wave, i.e. the outgoing scattered light. A material’s refractive index can be written as \( m(\lambda) = m_{\text{re}}(\lambda) + i m_{\text{im}}(\lambda) \), where \( m_{\text{re}} \) and \( m_{\text{im}} \) are real and imaginary parts of the refractive index. In strongly absorbing materials, the imaginary part is rather large in comparison to the real part. In strongly scattering materials, the real part is large compared to the imaginary part. When light is scattered by a molecule or by particles suspended in the atmosphere, the fraction of light that is scattered backwards towards the light source is called backscatter and has units of m⁻¹.
Figure 1.4: Scattering regimes as a function of scatterer radius $a$ and wavelength $\lambda$. The vertical green line indicates the 523 nm wavelength of the CALIOP lidar aboard the CALIPSO satellite. The square and star symbols along the green line indicate typical size ranges of atmospheric mineral dust particles (brown) and ice crystals (blue). Adapted from Wallace and Hobbs (2006).

The backscatter $\beta(\lambda, r)$ at wavelength $\lambda$ and distance $r$ from the light source can be expressed as

$$\beta(\lambda, r) = \Sigma_i n_i(r) \left. \frac{d\sigma_{sc}^i(\lambda, \Omega)}{d\Omega} \right|_{\Omega=180^\circ}$$

(1.3)

where $n_i(r)$ is the number of scatterers of type $i$ with scattering cross-sections $\sigma_{sc}^i(\lambda)$ at a distance $r$ from the light source, and where $d\sigma_{sc}^i(\lambda, \Omega)/d\Omega|_{\Omega=180^\circ}$ is the differential scattering cross-section, which is a measure of the likelihood that an incoming photon is scattered in the direction of the light source. Accounting for different scatterer types, the backscatter can also be written as

$$\beta(\lambda, r) = \beta_{\text{molec}}(\lambda, r) + \beta_{\text{part}}(\lambda, r)$$

(1.4)

where $\beta_{\text{molec}}$ and $\beta_{\text{part}}$ are the backscatter from molecules and from particles.

For a spherical scatterer of radius $a$, the size parameter $x = 2\pi a/\lambda$ is essentially the ratio of the radius $a$ to the wavelength $\lambda$ of the incident light. Figure 1.4 shows different scattering regimes as a function of wavelength and scatterer size. When the scatterer is much smaller than the wavelength ($x \ll 1$), the Rayleigh regime provides an approximate solution to Maxwell’s equations. The Rayleigh scattering cross-section is related to the wavelength by $\sigma_{\text{sc,Rayleigh}} \propto \lambda^{-4}$. When the scatterer is much larger than
Figure 1.5: Angular distribution of the intensities of light at wavelength of 0.5 \( \mu m \) scattered by spherical scatterers with radii (a) \( 10^{-4} \mu m \), (b) 0.1\( \mu m \), (c) 1\( \mu m \). The forward scattering is very large and has been scaled for the purpose of representation. From Liou (2002).

The wavelength \( x \gg 1 \), the path of the scattered light may be determined by means of geometric raytracing. When \( x \lesssim 1 \), the scattering cross-section may be calculated from Mie theory for spherical particles and by the T-matrix method for general particle shapes with one axis of rotational symmetry (Mishchenko et al., 2002).

Figure 1.5 illustrates the angular dependence of the scattered intensity of visible light (at \( \lambda = 500 \) nm) for spherical scatterers of different sizes. Panel (a) provides an example of Rayleigh scattering by a molecule, where approximately the same amount of light is scattered in forward and backward directions. The scattering takes increasingly place in the forward direction with growing particle size. A 1-\( \mu m \) particle, as shown in panel (c), scatters only a small fraction of the incoming light in the backward direction and most of it in the forward direction.

In a manner similar to Equation 1.3, the extinction can be written as the sum over all extinction cross-sections of scatters of type \( i \) present at a distance \( r \),

\[
\varepsilon(\lambda, r) = \Sigma_i n_i(r)\sigma_i^{\text{ext}}(\lambda) \tag{1.5}
\]

where \( \sigma_i^{\text{ext}} = \int_\Omega d\Omega \frac{d\sigma_i^{\text{ext}}}{d\Omega} \) is the total extinction (scattering plus absorption) cross-section. When light enters a medium at an intensity \( I_0(\lambda) \) and is backscattered by a scatterer positioned at a distance \( z \) into the medium, the optical depth of that medium along the path travelled by the incoming light is the integral over the extinctions along
the path, as expressed by Equation 1.2. The backscattered light traverses the medium a second time. Thus, the two-way transmission is given by

$$\frac{B(\lambda, r)}{I_0(\lambda)} = \exp^{-2\tau(\lambda, z)}.$$

(1.6)

where $B(\lambda, r)$ is the intensity of the backscattered light upon exiting the medium after the second traversal.

Cloud-aerosol lidar satellite observations. LIDAR (Light Detection And Ranging) is an optical measurement technique that allows range-resolving measurements of the state and composition of the atmosphere. A lidar instrument emits coherent high-intensity radiation at wavelength $\lambda$ and measures the backscatter $\beta(\lambda, r)$ from scatterers present at a distance $r$ from the instrument. It has been employed for purposes as diverse as distance measurements and the generation of digital surface models; the remote sensing of clouds and aerosols, their altitudes of occurrence, optical properties and composition; monitoring of water vapor and trace gases such as ozone and methane; measurement of wind speeds. Since lidar is an active remote sensing technique, measurements during nighttime are possible and even preferred, because they offer higher signal-to-noise ratios. Lidar allows measurements at the resolution of a few meters. It has a higher sensitivity to optically thin clouds and aerosols than radar and can differentiate between ice crystals and water droplets based on the amount of depolarization of the backscattered lidar signal.

Lidars have been operated ground-based, on aircraft, space shuttle and satellites. The Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) satellite shown in Figure 1.6 started operating in June 2006. It orbits Earth about 15.5 times a day in a near-polar orbit at an equatorial altitude of 705 km. CALIPSO carries the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) instrument aboard whose general setup is shown in Figure 1.7. CALIOP contains a Nd:YAG laser that emits simultaneous coaligned light pulses at 532 nm and 1064 nm in near-nadir direction. The pulses are 20 ns (about 6 m) long and have a repetition frequency of 20.16 Hz, so one profile is shot every 333 m along CALIPSO’s ground track. A beam expander (not shown) modifies the laser beam cross-section such that its footprint diameter on the ground measures about 70 meters. When the laser light encounters aerosol or a cloud on its way to the ground, part of the beam will be scattered back towards CALIPSO’s 1-meter diameter telescope. The light thus received is frequency-filtered to minimize
the solar background. The 532 nm channel then passes a polarization filter by which it is split into components parallel and perpendicular to the plane of the outgoing light. Both components are subsequently registered by a detector.

The range of intensities of backscattered light that CALIPSO can thus detect spans five orders of magnitude. In accordance with Wandinger (2005) and Young and Vaughan (2009), the power at wavelength $\lambda$ that is backscattered and detected from a target at a distance $r$ from the satellite can be expressed as

$$P(\lambda, r) = \eta P_0 c \Delta t A / (2r^2) \beta(\lambda, r) e^{-2\tau(\lambda, r)}.$$  \hspace{1cm} (1.7)

As explained in more detail in Wandinger (2005), $P(\lambda, r)$ is the product of (1) an instrument-specific term $\eta P_0 c \Delta t A / (2r^2)$, (2) the target-specific backscatter $\beta(\lambda, r)$, and an exponential term (3) that characterizes the optical depth $\tau$ of the medium between CALIOP and the target, as provided by Equation 1.6. With respect to the instrument-specific parameters, $P_0$ is the power of the emitted laser pulse, $c$ is the speed of light, $\Delta t = 20$ ns is the temporal pulse length and $c \Delta t = 6$ m the spatial pulse length, $A$ is the area of the receiver telescope and $\eta$ is the efficiency of the CALIOP detection system. Backscatter and extinction each have a molecular and a particulate component, as expressed in Equation 1.4 for backscatter. The molecular backscatter and extinction can be calculated as a function of atmospheric pressure and temperature. The variable measured at every distance $r$ from the lidar is $P(\lambda, r)$. Thus, two unknowns...
— particulate backscatter $\beta_{\text{part}}(\lambda, r)$ and particulate extinction $\varepsilon_{\text{part}}(\lambda, r)$ — are to be determined from the Lidar Equation 1.7, so one more condition is necessary. A target-specific backscatter-to-extinction ratio $\gamma(\lambda, r) = \varepsilon_{\text{part}} / \beta_{\text{part}}$, the so-called lidar ratio, is assumed for that purpose. For cirrus clouds, typical lidar ratios are on the order of 20–30 sr$^{-1}$. Explicit solutions to the Lidar Equation 1.7 are provided by e.g. Kovalev and Eichinger (2004).

The study presented in Chapter 3 makes use of CALIPSO measurements to investigate possible modifications of ice-cloud properties and ice-cloud occurrence caused by the stratospheric sulfate aerosol that formed following the eruption of the Nabro volcano in June 2011, the largest eruption since Pinatubo in 1991. Chapter 4 presents an ice-cloud climatology derived from CALIPSO measurements. The climatology is subsequently employed for the validation of ice nucleation schemes in the global climate-aerosol model ECHAM6-HAM2.
Chapter 2

Tropical Temperature and Precipitation Responses to large Volcanic Eruptions: Observations and AMIP5 Simulations

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Chapter 2 Surface anomalies following major volcanic eruptions

Abstract

We have examined tropical land mean surface air temperature and precipitation responses to the eruptions of El Chichón in 1982 and Pinatubo in 1991, as simulated by the 'atmosphere-only' GCMs (AMIP) of the Climate Model Intercomparison Project CMIP5, and compared them to three observational datasets. We statistically separated the El Niño-Southern Oscillation (ENSO) signal from the volcanic signal in all time series. Focussing on the ENSO signal, we found that the 17 investigated AMIP models successfully simulate the observed 4-months delay in the temperature responses to the ENSO phase, but simulate somewhat too fast precipitation responses during the El Niño onset stage. The observed correlation between temperature and ENSO phase (correlation coefficient of 0.75) is generally captured well by the models (simulated correlation of 0.71, ensemble means: 0.61 – 0.83). For precipitation, mean correlations with the ENSO phase are -0.59 for observations and -0.53 for the models, with individual ensemble members having correlations as low as -0.26. Observed, ENSO-removed tropical land temperature and precipitation decrease by about 0.35 K and 0.25 mm/day after the Pinatubo eruption, while no significant decrease in either variable was observed after El Chichón. The AMIP models generally capture this behavior, despite a tendency to overestimate the precipitation response to El Chichón. Scatter is substantial, both across models and across ensemble members of individual models. Natural variability thus may still play a prominent role, despite the strong volcanic forcing.

2.1 Introduction

Explosive volcanic eruptions have major impacts on the climate system, on time scales of a few months to a few years. Upon a strong eruption, sulfur dioxide (SO$_2$) may reach the lower stratosphere, where it is converted into aqueous sulfuric acid droplets that scatter shortwave and absorb infrared radiation and overall reduce the global mean surface air temperature. The volcanic aerosol may affect the formation of precipitation in several ways. Decreases in surface air temperatures lead to reduced evaporation and decreases in tropospheric column-integrated water vapor (Randel et al., 1996). Since precipitation is directly linked to evaporation, global mean precipitation decreases after a strong volcanic eruption (Robock and Liu, 1994). Once the stratospheric volcanic aerosol has been advected or sedimented into the upper troposphere, it may also influence cloud microphysical processes (Kübbeler et al., 2012; Cirisan et al., 2013), and consequently
Chapter 2 Surface anomalies following major volcanic eruptions

Precipitation. Stratospheric aerosol particles that form after a large volcanic eruption have typical stratospheric residence times on the order of a year. After returning to the troposphere, they may reside there for days up to a few weeks (SPARC, 2006) and during this time alter the lifetime and properties of clouds in the upper and middle troposphere. The response to radiative forcings is physically less constrained for precipitation than for temperature (Allen and Ingram, 2002). Accordingly, the signal-to-noise ratio of precipitation responses to volcanic eruptions is thought to be lower than for surface air temperature (Robock and Liu, 1994). In addition, a variety of dynamical feedback processes complicate matters further. The volcanic aerosol may induce vertical and horizontal heating gradients. These can affect stratospheric and tropospheric dynamical processes (see e.g. Graf et al. (1993), Ramachandran et al. (2000), Stenchikov et al. (2002)), which may affect the distribution of precipitation.

Observational studies indicate, for example, significant decreases in global and tropical-land-area precipitation following the June 1991 eruption of Mount Pinatubo (Trenberth and Dai, 2007; Gu and Adler, 2011). No significant reduction was observed after the eruption of El Chichón in April 1982, during which an estimated 40% of the amount of SO2 released during the Pinatubo eruption (18.5 ± 4 Mt) was emitted into the lower stratosphere (Gu et al., 2004; Krueger et al., 2008). Climate simulation studies suggest significant decreases in global and tropical-land-area mean precipitation following the eruptions of Toba about 74000 years ago (Robock et al., 2009; Timmreck et al., 2012) and of Pinatubo in 1991 (Broccoli et al., 2003), as well as based on composites of several volcanic eruptions (Robock and Liu, 1994; Joseph and Zeng, 2008; Schneider et al., 2009). Geoengineering has taken an interest in these violent events as observable, potential proxies to estimate the consequences of climate engineering via injection of sulfate into the stratosphere. In this overall context, it is of interest to examine the response of current generation climate models to the eruptions of Pinatubo and El Chichón.

Iles et al. (2013) studied the responses to volcanic eruptions simulated by atmosphere-ocean coupled climate models of the Coupled Model Intercomparison Project 5 (CMIP5). They found that the models simulate significant global precipitation reductions and that the reductions are largest in the tropics.

The aim of our study is to analyze how well the 17 atmosphere-only models of the CMIP5 project (‘AMIP5’) simulate the observed surface air temperature and precipitation responses to the eruptions of El Chichón (17°N) in 1982 and Mount Pinatubo (15°N) in 1991. The study is complementary to that of Iles et al. (2013) in two respects. First, in terms of the simulation data examined (AMIP instead of CMIP, i.e. using
prescribed, observation-based sea surface temperature data). Second, in that we remove the ENSO signal interfering with the volcano signal (see below) from both, observation and model data. Iles et al. (2013) removed it only from the observations and relied on statistical averaging out for the model data.

We focus on the El Chichón and Pinatubo events as they were by far the strongest eruptions after 1979, when satellite-based estimates of precipitation became available for global analysis. We focus on tropical land areas as precipitation reductions after low-latitude volcanic eruptions have been found to affect the tropical regions (20°N–20°S) in particular, which has been related to a weakening or contraction of the Hadley circulation (Robock and Liu, 1994; Trenberth and Dai, 2007; Schneider et al., 2009).

Identifying potential surface climate effects of volcanic aerosol is complicated by the fact that recent eruptions took place concurrently with warm phases of the El Niño-Southern Oscillation (ENSO). During such a warm phase, heat is transported from the ocean to the atmosphere in volcanically quiescent times, so surface air temperatures are enhanced while precipitation over tropical land regions decreases, as precipitation shifts from the land to the ocean (Trenberth et al., 2002). Thus, the influence of the ENSO partially masks the effects of the volcanic aerosol in surface temperature and precipitation time series. Over tropical land regions, for example, an El Niño phase and a strong volcanic eruption will each induce a precipitation reduction, while having counteracting effects on surface air temperatures. When studying the surface climate effects of large volcanic eruptions, it is necessary, therefore, to disentangle the volcanic and the ENSO influences.

In our AMIP5 data, we achieve this separation by applying the statistical lag-correlation/regression analysis method presented by Gu and Adler (2011), who investigated volcanic and ENSO signatures in the Global Precipitation Climatology Project (GPCP) product version 2.1 (Adler et al., 2003).

The objectives of our study are (1) to determine the time lags of the temperature and precipitation responses to changes in the ENSO phase based on observations and AMIP5 model simulations, (2) to investigate the observed and simulated sensitivity of the surface climate responses to the ENSO phase, and (3) to compare the magnitudes of the observed and the simulated posteruptive temperature and precipitation anomalies corrected for the ENSO contribution. We go beyond the work of Gu and Adler (2011) by investigating these questions for AMIP model simulations and for three different observational datasets of surface air temperature and precipitation.
Chapter 2 Surface anomalies following major volcanic eruptions

2.2 Data and methodology

2.2.1 Temperature observations

Observed 2-meter surface air temperatures were obtained from (1) the Global Historical Climatology Network and the Climate Anomaly Monitoring System (GHCN CAMS version 3.01, Fan and van den Dool (2008)), (2) the University of Delaware’s air temperature dataset (UDel version 3.01, Willmott and Matsuura (1995)), and (3) the University of East Anglia Climate Research Unit’s temperature dataset (CRU TS version 3.20, Harris et al. (2008)). These datasets contain global monthly station data interpolated to a $0.5^\circ \times 0.5^\circ$ grid. Because there are no satellite data of precipitation available prior to 1979, this year was chosen as the start year for the present study. We chose 2005 as the last assessed year for reasons explained below.

2.2.2 Precipitation observations

Observational data of tropical precipitation over land for years 1979–2005 are taken from three sources: (1) the Global Precipitation Climatology Project (GPCP version 2.2, Adler et al. (2003)), (2) the Climate Prediction Center’s Merged Analysis of Precipitation (CMAP version V1201, Xie and Arkin (1997)), and (3) the Precipitation Reconstruction over Land dataset (PRECL, version created 01/2011, Chen et al. (2002)). All three datasets provide gridded global monthly mean precipitation at $2.5^\circ \times 2.5^\circ$ spatial resolution. The GPCP and CMAP datasets are merged satellite and surface rain gauge estimates, while the PRECL dataset is rain-gauge-based only.

2.2.3 Model data

All simulated data come from the atmosphere-only 20th century CMIP5 simulations which have been run with observed sea-surface temperatures (SSTs) and sea ice concentrations (CMIP5 ‘experiment’ number 3.3, referred to as AMIP5) (Taylor et al., 2012). This ensures that the GCMs are subject to historically correct El Niño/ La Niña phases.

Table 2.1 provides an overview of the 17 models whose simulations we analyzed in this study. We considered only GCMs for which at least two ensemble members are available and which account for volcanic forcing. Ensemble sizes range from 2 to 6 members, as shown in table 2.1. Altogether, we have analyzed 64 ensemble members. For many of the models considered here, the atmosphere-only simulations have been run only for years after 1979 or before 2005. For that reason, our study is based on the years 1979–2005.
Different stratospheric aerosol forcing datasets have been used by the modeling groups, as CMIP5 does not provide emission data for volcanic aerosols. We have compared the three stratospheric aerosol optical depth (AOD) datasets most commonly used in the 17 models. The one by Sato et al. (1993) contains the stratospheric AOD at 550 nm. The Stenchikov et al. (1998) dataset provides stratospheric AOD in the 442–625nm solar band, while the stratospheric AOD of Ammann et al. (2003) is provided at 500 nm. The Ammann et al. (2003) and Stenchikov et al. (1998) datasets extend till December 1999 only. The stratospheric AOD time series of Sato et al. (1993) and Stenchikov et al. (1998) are very similar in the tropical mean, whereas clear differences exist in comparison to Ammann et al. (2003) whose peak tropical-mean stratospheric AOD is about 150% that of the former two datasets after both eruptions and whose stratospheric AOD decay times exceed those of the other two datasets.

2.2.4 Choice of posteruptive periods

We studied one-year posteruptive periods because the volcanic aerosol had a stratospheric residence time on the order of a year in the case of the El Chichón and Pinatubo events. June 1982–May 1983 (El Chichón) and August 1991–July 1992 (Mt. Pinatubo) were chosen as posteruptive periods to account for the time needed for the formation of aqueous H$_{2}$SO$_{4}$ aerosol droplets from the SO$_{2}$ that gathered in the lower stratosphere after the eruptions (SPARC, 2006). We repeated the analysis also for a two-year posteruptive period but obtained largely the same results (not shown in the following).

2.2.5 ENSO removal

Gu and Adler (2011) suggested that the El Niño/ La Niña signal may be removed in the tropics by making use of its strong correlation with tropical land 2-meter surface air temperatures or precipitation. We refer to this procedure as ‘ENSO removal’. Our study follows the ENSO removal method outlined by Gu and Adler (2011). Similar ENSO removal procedures have been applied by Robock and Mao (1995), Trenberth and Dai (2007), Chen et al. (2008), and Joseph and Zeng (2008).

In practical terms, we proceed as follows, for both our three observational and 64 AMIP5 model data sets. The Niño 3.4 index, which is the time series of monthly-mean SST anomalies averaged over the tropical Pacific (5°N–5°S, 120-170°W), was used to represent the ENSO phase. The index was computed from observed SSTs of Taylor et al. (2000). We interpolated all datasets bilinearly to a T63 grid (1.9° x 1.9°) for compa-
Table 2.1: CMIP5 models evaluated in this study

<table>
<thead>
<tr>
<th>Model index</th>
<th>Model</th>
<th>nlon × nlat</th>
<th>Ensemble members</th>
<th>Stratospheric aerosol</th>
<th>SSTs and sea-ice concentration</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>CESM1-CAM5</td>
<td>288 × 192</td>
<td>2</td>
<td>online</td>
<td>Hurrell et al. (2008)</td>
<td>Neale et al. (2010)</td>
</tr>
<tr>
<td>6</td>
<td>GISS-E2-R (p3)</td>
<td>144 × 90</td>
<td>6</td>
<td>online</td>
<td>Rayner et al. (2003)</td>
<td>Shindell et al. (2013, 2006)</td>
</tr>
<tr>
<td>8</td>
<td>MIROC5</td>
<td>256 × 128</td>
<td>2</td>
<td>online</td>
<td>Taylor et al. (2000)</td>
<td>Watanabe et al. (2010);</td>
</tr>
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<td></td>
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<td></td>
<td>Takemura et al. (2005)</td>
</tr>
<tr>
<td>11</td>
<td>MRI-CGCM3</td>
<td>320 × 160</td>
<td>3</td>
<td>online</td>
<td>Taylor et al. (2000)</td>
<td>Yukimoto et al. (2011)</td>
</tr>
</tbody>
</table>
Chapter 2 Surface anomalies following major volcanic eruptions

rability. Then, at each grid point, we constructed time series of surface air temperature anomalies. We computed the tropical mean as a 20°N–20°S area-weighted average, detrended the time series and removed the seasonal cycle by subtracting monthly climatologies based on volcanically largely unperturbed times (April 1979–March 1982, April 1985–March 1991, and April 1995–March 2005). We will refer to the detrended and deseasonalized time series as the filtered $T_{2m}$ time series subsequently. Pearson correlation coefficients of each filtered $T_{2m}$ time series with the Niño 3.4 index were determined. This was done using only the volcanically largely unperturbed time periods as provided above (April 1979–March 1982, April 1985–March 1991, and April 1995–March 2005). The filtered $T_{2m}$ time series were shifted with respect to the Niño 3.4 index by time lags of up to 12 months and a correlation coefficient was computed for each lag. The lag that corresponded to the (in absolute terms) maximal correlation coefficient was taken to be the mean response time of the filtered $T_{2m}$ time series to the ENSO phase over tropical land areas.

For each filtered $T_{2m}$ time series, we removed the ENSO signal by first performing a linear regression of the filtered time series, shifted by its lag with regard to the Niño 3.4 index. The regression provided an estimated filtered time series

$$\hat{T}_{2m}(t) = \beta_0 + \beta_1 \text{Niño3.4}(t - \text{lag}).$$

We obtained the ENSO-removed residual temperature by subtracting the estimate from the filtered $T_{2m}$ time series:

$$T_{2m}^{\text{ENSO removed}}(t) = T_{2m}(t) - \hat{T}_{2m}(t).$$

The regression coefficients were computed based on the volcanically unperturbed periods previously mentioned.

In an analogous manner, the lag-correlation/regression analysis was applied to the three precipitation observation time series and to all of the 64 ensemble members individually.

We note that non-linear ENSO effects remain poorly understood. By adopting a linear model for the relationship between the ENSO phase and filtered $T_{2m}$ (or precipitation), we assume that the ENSO phase is not significantly affected by climate responses to volcanic eruptions. As will become clear based on the datasets shown below, linear models appropriately describe the relationship between the Niño 3.4 index and filtered
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$T_{2m}$ (or precipitation) for the 1979–2005 time period considered in our study, as they can explain a large fraction of the variance induced in the filtered $T_{2m}$ and precipitation.

### 2.3 Results

#### 2.3.1 Lags and Correlations (tropical-land-area mean)

Shown in Figure 2.1 are the filtered temperature $T_{2m}$ for the different observational (red) and simulation (blue) datasets, and the Niño 3.4 index. There is some correlation between the filtered $T_{2m}$ and the ENSO time series, if the apparent lag between the two time series is properly taken into account. Comparing both time series in Figure 2.1, the temperature response apparently follows the ENSO phase with a time lag of a few months, both in the observations and in most ensemble members.

Figure 2.2 quantifies the correlation strength $R$ as a function of the number of months by which the respective temperature time series is shifted relative to the ENSO index. The lags are well defined, as each of the correlation functions is monotonously increasing towards a single maximum.

The lags of the observation-based time series are 4 months (GHCN CAMS and UDel) and 5 months (CRU TS). Those of the model-based time series range from 2 to 7 months with a mean of 4.3 months. Figure 2.2 also illustrates that for most models the scatter in time lag is small among the ensemble members.

The correlation coefficients between observed lag-shifted filtered temperature and the ENSO index are 0.74 (Udel) and 0.75 (GHCN CAMS and CRU TS). The simulated correlation coefficients range from 0.58 to 0.84 among the 64 ensemble members with a mean simulated correlation coefficient of 0.71. The strongest temperature-ENSO correlation is simulated by the CanAM4 model (ensemble mean: 0.83; model index 16), the weakest correlation by the MRI CGCM3 and FGOALS models (ensemble mean: 0.61 in each model; model indices 11 and 14, respectively).

Figure 2.3 illustrates the correlation between the Niño 3.4 index and the filtered precipitation. When comparing the onset times of ENSO events, such as the developing phase of the 1982/83 El Niño event at the beginning of 1982 or the 1997/98 El Niño developing phase at the beginning of 1997, it becomes clear that the observed and the simulated precipitation responses occur with a near-zero time lag. In several models, such as the MIROC5 and the IPSL-CM5A-LR (model indices 8 and 13, respectively), the precipitation over tropical land areas seems to respond more sensitively to changes
Figure 2.1: Filtered $T_{2m}$ and the Niño 3.4 index. Panel numbers 1–17 identify the models by the model indices provided in Table 2.1. The 'obs' panel presents the three observed filtered $T_{2m}$ time series. The gray bars indicate the eruptions of El Chichón (April 1982) and Pinatubo (June 1991). The time series variance $s^2$ (in K$^2$) is provided as an ensemble-mean for each model and as a mean over the observational datasets.
Figure 2.2: Shown on the y axis is the Pearson correlation coefficient R between filtered 2-meter surface air temperature \((T_{2m})\) time series over tropical land areas and the Niño 3.4 index for different time lags. The x axis provides the lags (in months). Positive lags indicate that the temperature curve reaches its maximum later than the Niño 3.4 curve. Different curves in each panel indicate the different ensemble members of the corresponding model. Panels as in Figure 2.1.
in the ENSO phase than is observed, in particular during El Niño onset phases. These models simulate precipitation minima before the ENSO index reaches its maximum value during the 1991/92 El Niño period. This may imply a too rapid shift of the simulated convective activity from the tropical land regions towards the ocean.

As shown in Figure 2.4, the observed and simulated filtered precipitation time series are significantly negatively correlated with the ENSO phase. Among the 64 ensemble members, the simulated lags range from -4 to +2 months with a mean lag of -1.8 months. The observed precipitation responds to the ENSO phase with a time lag of zero months (GPCP and PRECL) or one month (CMAP). This finding is in agreement with Gu and Adler (2011) who also determined a zero lag using an earlier version of the GPCP dataset. The observed mean precipitation reacts much faster to ENSO phase changes than $T_{2m}$ over tropical land regions, which is in agreement with Gu and Adler (2011). The latter study explained this observation by arguing that adjustments of the surface energy budget in response to ENSO phase changes take place more slowly than precipitation responses. The observed behavior (near-instantaneous precipitation response and delayed $T_{2m}$ response over tropical land regions) is successfully simulated by all 17 evaluated models, even though in most of the models the precipitation over tropical land areas responds too sensitively to a developing El Niño phase.

The correlation of the lag-shifted filtered observed precipitation with the Niño 3.4 index is -0.63, -0.61 and -0.54, respectively, in the GPCP, CMAP and PRECL datasets. The mean correlation over the 64 ensemble members is -0.53 (max/min values of -0.68 and -0.26), so the AMIP5 models tend to underestimate the ENSO-precipitation correlation strength over tropical land regions.

The relationship between lag-shifted filtered $T_{2m}$ and the ENSO phase can be assumed to be linear to a good approximation, as shown in Figure 2.5. The regression lines indicate that the observed $T_{2m}$ increases by about 0.16°C per unit of Niño 3.4 index. Most of the models simulate temperature responses in agreement with this observed ratio, whereas the CanAM 4 model (model index 16) clearly overestimates it. About 56% of the variance in the filtered observed $T_{2m}$ are explained by the linear regression on the Niño 3.4 index, as indicated by the coefficients of determination provided in Figure 2.5. Many models simulated temperature-ENSO relationships with $R^2$'s somewhat lower than observed, whereas CanAM 4 simulates a somewhat too high $R^2$ due to the strong coupling of its simulated $T_{2m}$ to the ENSO phase.

The observed negative relationship between precipitation and the ENSO phase can be estimated well by a linear regression line, as shown in Figure 2.6. According to
Figure 2.3: Same as Figure 2.1, but for filtered precipitation. The time series variance $s^2$ is provided in (mm/day)$^2$. 

\[ s^2 = 0.16 \quad (1) \]
\[ s^2 = 0.08 \quad (2) \]
\[ s^2 = 0.07 \quad (3) \]
\[ s^2 = 0.06 \quad (4) \]
\[ s^2 = 0.06 \quad (5) \]
\[ s^2 = 0.06 \quad (6) \]
\[ s^2 = 0.10 \quad (7) \]
\[ s^2 = 0.07 \quad (8) \]
\[ s^2 = 0.08 \quad (9) \]
\[ s^2 = 0.07 \quad (10) \]
\[ s^2 = 0.14 \quad (11) \]
\[ s^2 = 0.07 \quad (12) \]
\[ s^2 = 0.06 \quad (13) \]
\[ s^2 = 0.11 \quad (14) \]
\[ s^2 = 0.08 \quad (15) \]
\[ s^2 = 0.08 \quad (16) \]
the three observational datasets of precipitation, the filtered precipitation is reduced by 0.1 mm/day per unit of Niño 3.4 index. The majority of the models successfully simulate a similar ratio. About 35% of the observed variance in filtered precipitation is explained by the regression. The simulated $R^2$s range from 7–46% among the ensemble members.

### 2.3.2 ENSO-removed Temperature and Precipitation (tropical-land-area mean)

Figure 2.7 provides the $T_{2m}^\text{ENSO\ removed}$ time series, i.e. filtered $T_{2m}$ after the ENSO signal removal by lag-correlation/regression as explained above. The time series are 12 months shorter at both ends as compared to before the ENSO removal (Figure 2.1) because our approach for determining the time lags allows for shifting by up to ±12 months.

As can be seen, removing the ENSO contributions significantly reduced the variance of the filtered $T_{2m}$ time series for all models and in all observational datasets, as expected. The mean variance of the three $T_{2m}^\text{ENSO\ removed}$ observational datasets was reduced from 0.08 to 0.05 K$^2$, for example.

Focussing now on our original goal, the impact of the El Chichón and Pinatubo eruptions as seen in appropriately treated time series, mean temperature reductions of about
Figure 2.5: Regression of lag-shifted filtered $T_{2m}$ on the Niño 3.4 index. To identify the models, the numbers in parentheses indicate the model indices as provided in Table 2.1.
Figure 2.6: Regression of lag-shifted filtered tropical mean precipitation over land on the Niño 3.4 index. To identify the models, the numbers in parentheses indicate the model indices as provided in Table 2.1.
Figure 2.7: Residual ENSO-removed surface air temperature ($T_{2m}^{\text{ENSO \ removed}}$). Model indices, volcanic eruption times and time series variances are indicated like in Figure 2.1.
Figure 2.8: Residual ENSO-removed precipitation. Model indices, volcanic eruption times and time series variances are indicated like in Figure 2.1.
0.4 and 0.6 K are observed over tropical land areas after the El Chichón and Pinatubo eruptions in late 1982 and the second half of 1992, respectively, in agreement with Gu and Adler (2011) (their Figures 8b, 3b).

The variances in the filtered precipitation time series were likewise significantly reduced by the ENSO removal for all models and the observational datasets, as shown in Figure 2.8. The mean precipitation observed over tropical land areas is clearly reduced by up to 0.3 mm/day following the Pinatubo eruption, whereas the observed precipitation response to the eruption of El Chichón is more ambiguous, in agreement with Gu and Adler (2011) (their Figures 7b, 2b).

Figure 2.9 shows the ranges of the $T_{2m}^{\text{ENSO removed}}$ and precipitation anomalies after ENSO removal for each ensemble member. The whiskers indicate the maximum and minimum anomalies.

Before determining the significance of the observed and simulated post-eruptive $T_{2m}^{\text{ENSO removed}}$ and precipitation reductions, we found that the residual $T_{2m}^{\text{ENSO removed}}$ and precipitation time series are significantly autocorrelated at a lag of one month. To remove the autocorrelation, we “prewhitened” the time series by transforming them according to $y'_t = (y_t - r_1y_{t-1})/(1 - r_1)$ (von Storch and Navarra, 1993; Wang and Swail, 2001), where $r_1$ is the lag-1 autocorrelation coefficient. A two-sided Mann-Whitney test was performed on the prewhitened time series of residual ensemble-mean $T_{2m}^{\text{ENSO removed}}$ and precipitation anomalies to test the null hypothesis that the mean of the post-eruptive monthly anomalies is not significantly different from the mean of the control period at the 95% level.

The anomalies shown in Figure 2.9 are means over the above 12-months post-eruptive periods. As shown in Figure 2.9, the observed $T_{2m}^{\text{ENSO removed}}$ and ENSO-removed precipitation dropped significantly by about 0.35 K and 0.25 mm/day after the Pinatubo eruption. This is in good agreement with the Niño-3.4-removed filtered surface air temperature and precipitation responses obtained by Gu and Adler (2011) (their Figures 8b and 7b) based on an earlier version of the GPCP dataset. The observed $T_{2m}^{\text{ENSO removed}}$ and precipitation reductions of about 0.17 K and 0.05 mm/day in the year after the El Chichón eruption are not statistically significant.

The AMIP5 models generally simulate $T_{2m}^{\text{ENSO removed}}$ and ENSO-removed precipitation responses in agreement with the magnitude of the observed responses. There is, however, a tendency for overestimating the observed (non-significant) precipitation response to the El Chichón eruption over tropical land areas. Also, there is large scatter in the simulated anomalies, both across models and across ensemble members within a
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Figure 2.9: Simulated anomalies in $T_{2m}^{\text{ENSO removed}}$ and ENSO-removed precipitation after the El Chichón and Pinatubo eruptions, averaged over June 1982–May 1983 and August 1991–July 1992, respectively. Ensemble-mean anomalies are marked by a triangle when significantly (at the 95% level) below the model’s control-period mean, or by a circle otherwise. Ensemble minimum and maximum anomalies are marked by the upper and lower whisker edges. Gray shadings denote the ranges of observed posteruptive anomalies.
single model. The latter is interesting as it may point to a still significant role of natural variability even in the presence of strong volcanic forcing. Of course, other interpretations are possible as well, from model deficiencies to insufficient quality of the ENSO removal.

Figure 2.10 shows the simulated and observed anomalies of all ensemble members. The mean and standard deviation of the simulated surface temperature and precipitation anomalies, taken over all ensemble members of all 17 models, are \(-0.08 \pm 0.11\) K and \(-0.12 \pm 0.09\) mm/day for the El Chichón eruption, and \(-0.29 \pm 0.11\) K and \(-0.26 \pm 0.10\) mm/day for Pinatubo. This corresponds to signal-to-noise ratios of 0.7 and 1.3 for the anomalies in $T_{2m}^{\text{ENSO removed}}$ and precipitation in the case of El Chichón eruption, and of 2.6 for both the $T_{2m}^{\text{ENSO removed}}$ and the precipitation anomalies for Pinatubo.

The ranges (max - min) of the simulated $T_{2m}^{\text{ENSO removed}}$ and precipitation anomalies over all ensemble members and models are 0.52 K and 0.40 mm/day (El Chichón) and 0.49 K and 0.51 mm/day (Pinatubo). The large scatter in the simulated temperature and precipitation anomalies among the 64 ensemble members illustrates the small ratio of volcanic signal to model internal variability (and to model-to-model variability) in the AMIP5 simulations.

It is interesting to note that there is no apparent relation between the strength of the $T_{2m}^{\text{ENSO removed}}$ response and that of the ENSO-removed precipitation response.

The Pinatubo eruption was 2–3 times larger than the eruption of El Chichón in terms of the stratospheric SO$_2$ input (SPARC, 2006). Figure 2.10 suggests that the stratospheric AOD increase and the reduction in evaporation following the El Chichón eruption were not large enough to cause a response in $T_{2m}^{\text{ENSO removed}}$ and ENSO-removed precipitation beyond the level of natural variability.

### 2.4 Conclusions

We have investigated surface air temperature and precipitation responses to the eruptions of El Chichón in April 1982 and Mount Pinatubo in June 1991 over tropical land areas, as simulated by 17 'atmosphere-only' GCMs of the Climate Model Intercomparison Project CMIP5.

The objectives of our study were to validate the time lags and coupling strengths of the simulated surface climate responses to changes in the ENSO phase and to validate the observed and the simulated posteruptive temperature and precipitation anomalies over tropical land areas by comparison to observed temperature and precipitation.
Figure 2.10: Simulated anomalies in ENSO-removed surface air temperature ($T_{2m}$ ENSO removed) and ENSO-removed precipitation anomalies of the 64 ensemble members, averaged over June 1982–May 1983 (El Chichón) and August 1991–July 1992 (Pinatubo). Observational values are marked by bold edges. Circles indicate a significant reduction in $T_{2m}$ ENSO removed and ENSO-removed precipitation; triangles (squares) mark significant decreases in $T_{2m}$ ENSO removed (ENSO-removed precipitation) only. Diamond symbols were chosen when there was no significant post-eruptive decrease in any of the two variables. Significance was tested at the 95% level in all cases. The marker colors indicate the associated model in accordance with the color coding used in Figure 2.9.
Focussing on the responses over tropical land regions, we found that:

- All models successfully simulate surface air temperature responses delayed by a few months relative to the ENSO phase (on average 4.3 months), which agrees well with the observed 4-5 months delay.

- The strong positive correlation observed between mean temperatures and the ENSO phase (correlation coefficient of 0.75) is generally captured well by the models (simulated correlation of 0.71). There is, however, considerable scatter in the simulated correlation strength across models (ensemble means of 0.61–0.83).

- The observed precipitation response lags the ENSO phase by 0–1 months. Most of the models appear to simulate a somewhat too fast precipitation response during the El Niño onset (mean simulated lag of $-1.8$ months). This may be related to a too rapid shift of simulated convective activity towards the ocean.

- The models tend to underestimate the observed correlation strength between precipitation and ENSO phase (mean correlation of $-0.59$). The mean simulated correlation coefficient is $-0.53$. Simulated correlations of ensemble members range as low as $-0.26$.

- The observed relationship between ENSO phase, as measured by the Niño 3.4 index, and temperature (or precipitation) can be considered linear to a reasonably good approximation for 1979–2005. The models successfully capture this linearity, though in the case of temperature typically at lower $R^2$s.

- The observed mean temperature and precipitation increase by 0.16$^\circ$C and 0.1 mm/day per unit of Niño 3.4 index. Many but not all models simulate temperature and precipitation sensitivities in agreement with this finding.

- Observed ENSO-removed temperature and precipitation decreased by about 0.35 K and 0.25 mm/day after the Pinatubo eruption, whereas no significant decrease in either variable was observed after El Chichón. The models generally capture this behavior, even though with large scatter. They appear to somewhat overestimate the precipitation response to El Chichón.

- The stratospheric AOD increase and associated reduction in evaporation after El Chichón eruption seem to not have been large enough to result in temperature or precipitation responses beyond the level of natural variability.
Obviously, the present study is only a first step. An area we did not touch upon is the model physics behind the good or bad agreement between observations and one particular model. Or, similarly, the reason for the considerable spread across ensemble members for at least some models. Another question concerns the quality of the ENSO removal. One possible way forward here could be a dedicated modelling study: perform two sets of atmosphere-only model simulations that differ only in the presence or absence of Pinatubo-like eruptions and check whether the ENSO removal procedure applied here can indeed retrieve the difference signal of the two sets of simulations.
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Chapter 3

Did the 2011 Nabro eruption affect the optical properties of ice clouds?

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Chapter 3 Did the 2011 Nabro eruption affect the optical properties of ice clouds?

Abstract

The eruption of the Eritrean Nabro volcano in June 2011 was the largest eruption since Mount Pinatubo in June 1991. The Nabro volcano emitted 1–1.5 Mt of sulfur dioxide into the lower stratosphere which resulted in a significant rise in the stratospheric sulfate aerosol burden in the months following the eruption. We have analyzed backscatter and extinction from ice clouds, as measured by the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) satellite between June 2006 and May 2014, to assess if volcanic aerosol produced by the Nabro eruption had affected ice clouds. We found no significant modifications of either of ice-cloud optical properties (i.e. total backscattering and extinction), occurrence frequencies or residence altitudes on a global scale. Using the analyzed optical properties as indicators of post-eruptive ice-cloud radiative forcing modifications, we find that the eruption had no significant volcanic aerosol-ice cloud radiative effect. Our results suggest that the investigated optical properties of ice and cirrus clouds are at most weakly dependent on the sulfate droplet number density and size distribution.

3.1 Introduction

Cirrus clouds play an important role in the radiative budget of the Earth (Liou, 2002). They are thought to have a positive cloud radiative effect (Chen et al., 2000): Their cloud tops are at cold temperatures, so they emit less infrared radiation than clouds at lower altitudes or the surface. Moreover, the greater the cirrus optical thickness, the more terrestrial infrared radiation is emitted back to the ground. On the other hand, thicker cirrus clouds will scatter more solar radiation back to space. This cooling effect is outweighed by the aforementioned warming effects at typical cirrus optical thicknesses and altitudes (Corti and Peter, 2009).

Cirrus clouds form when a sufficiently moist air parcel is lifted to altitudes at which freezing is possible. Uplifts can occur in synoptic weather systems, in convection, in the tropical tropopause region (cold trap) and over mountainous terrain, as outlined in Sassen (2002). In the case of contrail cirrus, aircraft exhausts provide the water vapor that is necessary for ice crystals to form. Tropospheric ice crystals can form in cirrus clouds in the upper troposphere at temperatures below $T_{hom} \approx -40^\circ$C. Liquid droplets can freeze homogeneously only below $T_{hom}$. At warmer temperatures, freezing is possible in mixed-phase clouds at intermediate altitudes only via formation of ice-
germs on the surfaces of insoluble aerosol particles in a process called heterogeneous nucleation (Pruppacher and Klett, 1997). Deposition nucleation is a heterogeneous nucleation pathway that can take place at temperatures below $T_{\text{hom}}$. Depending on an air parcel's initial humidity, Wiacek et al. (2010) outlined possible histories of cirrus- and mixed-phase-clouds-forming air parcels, and the relevance of homogeneous and heterogeneous nucleation therein.

In the context of this work, we study both cirrus clouds (including cirrostratus, cirrocumulus, convective anvil cirrus) and the iced tops of mixed-phase clouds, i.e. the upper regions of nimbostratus and cumulonimbus clouds at temperatures $T < -40^\circ$C. The global annual mean occurrence of Nimbostratus (Ns) and Cumulonimbus (Cb) clouds are 5-9\% over land and 5-11\% over ocean, while Cirrus (Ci), Cirrostratus (Cs) and Cirrocumulus (Cc) have a global annual mean occurrence of 6-22\% over land and 6-14\% over ocean (Raschke et al., 2005; Eastman and Warren, 2013). Adopting mean values of 7\% and 14\% for Ns+Cb and Ci+Cs+Cc over land (and of 8\% and 10\% over ocean), the fraction of iced tops of mixed-phase clouds relative to all ice clouds is 33\% over land and 44\% over ocean.

We sample the CALIPSO cloud observations based on a temperature criterion. Homogeneous freezing of micrometer-size droplets of pure water occurs at about $-38^\circ$C (Koop et al., 2000). To account for freezing point depression effects caused by atmospheric background concentrations of sulfate ions, we take $-40^\circ$C as our ice-cloud detection threshold. To test the sensitivity of our results with respect to this criterion, we repeated the analysis with $-50^\circ$C, $-60^\circ$C and $-70^\circ$C thresholds.

Stratospheric sulfate aerosols may have the potential to modify the microphysical and radiative properties, occurrence frequencies and altitudes of ice clouds (Sassen, 1992; Jensen and Toon, 1992; Wang et al., 1995; Song et al., 1996; Kühbeler et al., 2012). The objective of this study is to investigate whether sulfate aerosols from the 2011 eruption of the Nabro volcano have affected the optical properties (i.e. the total backscatter and extinction) of ice clouds on a global scale.

The Nabro volcano (13.37°N, 41.70°E) is located in the Afar Depression, part of the East African Rift located in the Eritrean-Somalian border region. The eruption started on 12 June 2011 at around 21 UTC (Vernier et al., 2013) and lasted for about a month. As reported by Theys et al. (2013), about 4.5 Mt of sulfur dioxide ($\text{SO}_2$) were released during this time period, of which an estimated 1 Mt reached the lowermost stratosphere (N. Theys, personal communication). Robock (2013) provides an estimate of 1.5 Mt for the stratospheric $\text{SO}_2$ input following the eruption. The plume was rich in $\text{SO}_2$ and water
vapor, and poor in ash if any (Clarisse et al., 2012; Theys et al., 2013; de Vries et al., 2014). A large fraction of the total SO$_2$ release occurred during the first 15 hours of the eruption with injection heights between 15-18 km (Theys et al., 2013). SO$_2$ was injected directly to or above the tropopause region during this initial eruption and on 16 June 2011 (Fromm et al., 2014). Parts of the initial plume were transported towards East Asia in the upper troposphere (Sawamura et al., 2012; de Vries et al., 2014). Bourassa et al. (2012, 2013) have claimed that the plume was advected into the lower stratosphere by deep convection linked to the Asian monsoon. Vernier et al. (2013) and Fromm et al. (2013, 2014) argued that the plume was injected directly into the stratosphere instead. Clarisse et al. (2014) confirmed that lower parts of the initial plume underwent uplift in the Asian monsoon circulation, while Fairlie et al. (2014) asserted that this uplift was not due to monsoon convection but can be explained by quasi-isentropic flow alone.

Following the Nabro eruption, the stratospheric aerosol load increased mainly over Asia in the first month following the onset of the eruption (Bourassa et al., 2012). Figure 3.1 shows the stratospheric aerosol-to-molecular backscatter ratio from the first half of June 2011 to the second half of December 2011 based on nighttime backscatter measurements of the CALIPSO satellite. A stratospheric aerosol plume formed within weeks after the eruption onset, with the peak zonal-mean aerosol optical depth at about 20 to 25°N. The feature observed near 10–12 km altitude North of 45°N in June and the first half of July is likely the plume from the Grimsvötn eruption in May 2011. Figure 3.2 shows the stratospheric aerosol optical depth at 532 nm above 15 km from June 2006 to May 2014 based on CALIPSO observations. By mid-August 2011, the Nabro plume had spread over the entire Northern hemisphere (NH; Figures 3.1 and 3.2), and by end 2011 most of the stratospheric aerosol from the Nabro eruption had returned to the troposphere by stratosphere-to-troposphere exchange of air and sedimentation.

The Nabro eruption was likely the largest eruption since Mount Pinatubo (16°N) in June 1991 in terms of its stratospheric SO$_2$ injection. During the Pinatubo eruption, about 18 ± 4 Mt of SO$_2$ (Guo et al., 2004) and 5–6 km$^3$ dense-rock equivalent of volcanic ash (Wiesner et al., 2003) were released into the lower stratosphere. The Pinatubo eruption induced stratospheric warming and tropospheric cooling responses (McCormick et al., 1995). Free and Angell (2002) have examined the effects of volcanic eruptions on the vertical temperature structure of the troposphere using radiosonde data. They showed for the tropics that a tropospheric cooling followed the Pinatubo eruption which was maximal in the mid to upper troposphere, while the El Chichón (17°N, April 1982) and Agung eruptions (8°S, March 1963) exhibited a smaller and vertically more
Figure 3.1: Zonal mean backscatter ratio (aerosol to molecular) of the Nabro plume between 0 – 62° N and 8–20 km altitude from June to December 2011 from CALIOP Level 1 nighttime data. The red curve shows the zonal mean tropopause altitude from GEOS-5 data.
evenly distributed tropospheric temperature response. How a volcanic eruption affects the properties and abundance of ice particles may depend on the upper tropospheric temperature response to the eruption. As some eruptions may potentially induce significant temperature responses, our results may not be generalizable to arbitrary volcanic eruptions.

### 3.2 Potential effects of volcanic aerosols on ice clouds

*Sassen* (1992) and *Sassen et al.* (1995) have reported cirrus clouds with ice crystal number densities as high as 600 per liter that were observed over Kansas in December 1991 with a ground-based lidar in the aftermath of the June 1991 eruption of Mount Pinatubo. They also suggested that the upper parts of two observed cirrus clouds, which were at temperatures between $-40^\circ$ and $-50^\circ$C, i.e. below the homogeneous freezing temperature of water, consisted mainly of supercooled aqueous sulfuric acid droplets based on depolarization data. The authors explained this observation with a freezing-point depression that occurred in cloud droplets after an influx of volcanic aerosols into the cirrus-forming region. In the context of their study, a freezing-point depression is the process of reducing the freezing point of water by adding a solute, sulfuric acid, to it. The authors concluded that their observations provided evidence of a so far unobserved volcanic-aerosol cirrus-cloud climate feedback mechanism. *Sassen* (1992)
also suggested that an influx of sulfate droplets into the upper troposphere might increase the abundance of cirrus clouds, because stratospheric sulfuric acid solution droplets that enter a cirrus-forming air mass after a volcanic eruption are typically larger than in volcanically quiescent times. The freezing probability of solution droplets increases with the droplet volume according to classical nucleation theory and in accordance with numerous laboratory measurements (Pruppacher and Klett, 1997). Larger droplets are, therefore, more likely to freeze. Moreover, the larger a solution droplet and its sulfuric acid concentration, the lower the relative humidity that is required for the droplet to be in thermodynamic equilibrium with the gas phase, as described by the Kelvin and Raoult terms in the Köhler equation (Pruppacher and Klett, 1997). Thus, the authors further suggested that cirrus clouds might occur at lower relative humidities after a strong volcanic eruption. Finally, they proposed a post-eruptive increase in the cirrus lifetimes, because a greater abundance of large sulfate droplets might cause more and smaller ice crystals to form which would sediment less rapidly.

Wang et al. (1995) analyzed Stratospheric Aerosol and Gas Experiment (SAGE) II observations of subvisible tropical tropopause cirrus clouds following the El Chichón eruption in 1982. They found that clouds having high extinction coefficients become less frequent after the eruption, while clouds with low extinction coefficients become more frequent. The authors pointed out that, according to Mie theory, the cloud extinction coefficient decreases with increasing aerosol number densities if the effective cloud particle radii are below 0.8 µm. They concluded that their cirrus cloud observations consist of particles of effective radii smaller than 0.8 µm. They concluded further that the increase in number density of volcanic aerosol had induced this reduction in the cloud particle effective radii, because a greater number of solution droplets had been competing for the same amount of available water vapor after the eruption.

Song et al. (1996) analyzed outgoing longwave radiation data from the NOAA Climate Analysis Center, and SAGE II and ground-based lidar observations. They found high-level cloudiness to increase with the global stratospheric aerosol load, particularly in the midlatitudes, and concluded that volcanic aerosol can significantly modify high-level cloudiness and thereby affect global climate. To explain their observations, the authors suggested that the post-eruptive enhancement in solution droplet number densities increased the abundance of cloud condensation nuclei in the upper troposphere in post-eruptive times, which may have enhanced upper level cloudiness.

Massie et al. (2003) examined Halogen Occultation Experiment (HALOE) and SAGE II observations of tropical tropopause cirrus. They found decreases in the occurrence
frequency and the extinction of cirrus clouds after the 1991 eruption of Mount Pinatubo, in agreement with Wang et al. (1995). The authors hypothesized that increases in ice nuclei number densities or changes in the strength or occurrence frequency of deep convective events might explain their observations. Moreover, they analyzed anomalies of radiosonde temperatures near the tropical tropopause together with SAGE II cirrus extinction measurements in order to investigate whether tropopause temperature changes as observed after the Pinatubo eruption can explain the post-eruptive decrease in cirrus extinction. Based on their analysis, they excluded tropopause temperatures as a possible explanation of the observed post-eruptive reduction in cirrus extinction.

Luo et al. (2002) analyzed three datasets that were derived from passive satellite instruments for evidence of large-scale modifications of cirrus cloud amounts or optical properties, and found no such effects. They concluded that “previous studies showing some changes are probably isolated local effects”. Rolf et al. (2012) examined ground-based lidar data of a cirrus cloud situated in a volcanic plume which originated from the 2010 eruption of the Icelandic Eyjafjallajökull volcano. Running a microphysical box model along back trajectories, they concluded that the observed cirrus cloud must have formed by heterogeneous nucleation on the volcanic ash particles of the plume.

Turning to modelling studies, Jensen and Toon (1992) performed simulations on the effects of volcanic sulfate aerosols on cirrus clouds. They found that ice crystal number densities can increase by up to a factor of 5 when an air mass rich in sulfate droplets of stratospheric origin mixes with a tropospheric cirrus-forming air parcel. According to their estimates, this volcanic aerosol-cirrus cloud effect may amount to a net surface warming of up to 8 W/m². The authors stressed the sensitivity of their results to a number of assumptions made, and the need of further observational studies to constrain the model uncertainties. According to Jensen et al. (1994), the microphysical properties of cirrus clouds are, however, primarily sensitive to temperature and cooling rates of the cirrus-forming air mass, and at most weakly related to the sulfate droplet number density.

Lohmann et al. (2003) simulated increases in ice crystal number density by about 50–100% in the tropical tropopause region following the Pinatubo eruption. They found that ice crystal number densities tended to be enhanced in regions where solution droplet number densities were low. Moreover, their results agreed with Luo et al. (2002) in that the sulfate aerosols of the simulated Pinatubo eruption have no significant cloud radiative effect.
Kübbeler et al. (2012) studied effects of stratospheric sulfate geoengineering on cirrus clouds and found that injections of 5 Mt SO$_2$/a caused reductions in ice crystal number densities by 5-50%. The simulated increase in stratospheric aerosol load warmed the upper troposphere and lower stratosphere (UTLS), because the aqueous sulfuric acid solution droplets absorb and re-emit terrestrial radiation and thereby warm the surrounding air masses. The UTLS warming in turn stabilized the middle and lower troposphere and thus reduced vertical wind velocities in cirrus-forming air parcels. This caused homogeneous ice nucleation rates to decline, resulting in cirrus clouds that were substantially thinner optically. The resulting radiative cooling effect made up 60% of the overall net radiative effect of the employed geoengineering scheme. Cirisan et al. (2013) studied stratospheric sulfate geoengineering effects on cirrus forming air parcels in the Northern midlatitudes from injections of 2–10 Mt SO$_2$/a. They found increased ice crystal number densities in cirrus clouds that formed in air masses strongly affected by stratospheric air, and reduced ice crystal number densities in air masses only mildly affected by stratospheric air. The authors report a midlatitude cirrus radiative effect averaging out to less than 1% of the overall net radiative effect of their geoengineering scheme. Comparing to Kübbeler et al. (2012), the study of Cirisan et al. (2013) employs a more comprehensive microphysical scheme, but focuses on the Northern midlatitudes only and does not consider feedbacks, such as upper tropospheric temperature and humidity changes.

3.3 CALIPSO-based ice-cloud statistics

Many passive satellite instruments are known to sample only the optically thick ice clouds and to miss optically thin ones (Stubenrauch et al., 2013). The CALIOP lidar on the CALIPSO satellite is able to detect even subvisible ice clouds. Based on the CALIOP backscatter and extinction signals, the residence altitudes, optical thicknesses and occurrence frequencies of ice clouds can be calculated, which determine the cloud radiative effect (Corti and Peter, 2009). Therefore, if significant modifications of the shape or position of the frequency distribution of lidar backscatter or extinction from clouds were observed in post-eruptive seasons, this would suggest a cloud radiative effect of the volcanic aerosol. CALIOP is one of the most suitable instruments currently available for investigating the global radiative effect of volcanic aerosols on ice clouds. CALIPSO has been probing Earth’s atmosphere nearly continuously since June 2006. It provides vertical profiles of backscatter and depolarization at 532 nm from particulates suspended
in the troposphere and lower stratosphere during 15 orbits a day (Winker et al., 2007, 2009). The CALIPSO level 2 retrieval algorithms have been employed on the profiles to discriminate cloud and aerosol particles (Vaughan et al., 2009). We used the retrieved CALIPSO level 2 cloud profile product version 3 provisional (L2 CPro) (Anselmo et al., 2007) to analyze nighttime measurements of particulate backscatter from 13 June 2006 to 31 May 2014. The data are provided at a resolution of 5–80 km along the ground track and 60 m vertically.

The 532 nm backscatter signal-to-noise ratios are worse during daytime because of the greater number of solar photons reaching the receiver telescope. Therefore, some thinner cirrus clouds may remain undetected by CALIPSO during daytime (Winker et al., 2009). Apparent day-/nighttime differences in cirrus cloud occurrence frequency in the L2 CPro data product may result either from actual differences in occurrence frequency or from the poorer daytime signal-to-noise ratio. It is not possible to compare day- and nighttime cirrus occurrence frequencies based on merely the L2 CPro data without being affected by the daytime low bias (M. Vaughan, personal communication). Therefore our study focuses on the nighttime data only.

We also use atmospheric temperature data from the Goddard Earth Observing System Model data assimilation system, version 5 (GEOS-5) of NASA’s Global Modeling and Assimilation Office. We filter the backscatter and extinction data based on the cloud-aerosol discrimination (CAD) score (Liu et al., 2009, 2010) which provides a measure of the level of confidence in the result of the cloud-aerosol discrimination algorithm of Vaughan et al. (2009). The CAD score distinguishes clouds from aerosol based on five-dimensional cloud and aerosol distributions, $p_{\text{cloud}}$ and $p_{\text{aerosol}}$, that were developed based on four months of CALIOP observations and whose dimensions are the attenuated backscatter at 532 nm, the ratio of attenuated backscatter at 532 nm and at 1064 nm, the depolarization ratio, latitude and altitude. The CAD score is defined as

$$\frac{p_{\text{cloud}} - p_{\text{aerosol}}}{p_{\text{cloud}} + p_{\text{aerosol}}} \times 100$$

where $p_{\text{cloud}}$ and $p_{\text{aerosol}}$ take values in the range of 0 to 1. The CAD score values range from 0 to 100 for clouds and from 0 to −100 for aerosol. If there is no overlap between the 5D cloud and aerosol distributions, clouds and aerosol can be distinguished from each other on the highest confidence level (CAD score =100 for clouds, CAD score = −100 for aerosol). Absolute CAD score values larger than 70, between 50 and 70, between 20
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and 50, and smaller than 20 correspond to high/medium/low/no confidence in the cloud-aerosol discrimination according to ASDC.

We filter the backscatter and extinction measurements such that any measurements with CAD score below 100 are discarded from our analysis. While (even at CAD score =100) the CAD algorithm may not work fully reliably when discriminating mineral dust, volcanic ash or smoke from cirrus clouds (ASDC), it performs well in distinguishing cirrus clouds from sulfate plumes based on their depolarization characteristics: sulfate droplets have a depolarization ratio of close to zero because of their sphericity, whereas ice crystals in clouds at $T \leq -40^\circ C$ typically produce lidar depolarization ratios of about $0.35 \pm 0.1$ (Sassen and Benson, 2001). The good performance of the CAD algorithm in the case of sulfate-versus-cirrus discrimination is illustrated by the CALIPSO browse images of the 2011 Nabro plume (e.g. Browse Images, 2011). The plume was rich in SO$_2$ but contained very little if any ash (Clarisse et al., 2012; Theys et al., 2013; de Vries et al., 2014). Based on CALIPSO browse image inspections of the Nabro plume and on the negligible role of ash in the plume, we concluded that our results are not affected by a significant number of instances of volcanic-aerosol misclassification as cloud.

Polar stratospheric clouds (PSC) are generally not contained in the L2 CPro dataset (ASDC, 2013). In some cases, we found the PSC removal incomplete though, so we screened the remaining PSC pixels by removing all backscatter measurements taken at altitudes above 14 km at latitudes polewards of 65°.

3.4 Results

As shown in Figure 3.1, a large fraction of the stratospheric aerosol that formed following the Nabro eruption returned to the Northern hemispheric troposphere by the end of 2011. Post-eruptive modifications of ice cloud properties would, therefore, mostly be expected in the NH and within the first half year after the eruption.

3.4.1 Zonal-mean ice–cloud occurrence frequencies

Figure 3.3 shows monthly-mean zonal-mean nighttime ice occurrence frequencies from the start of the operation period of the CALIPSO satellite in June 2006 to May 2014 for several latitude bands. In each panel, the black curve represents ice occurrence frequencies below a threshold temperature of $-40^\circ C$, i.e., the ratio of the number of profiles which contain at least one in-cloud measurement below $-40^\circ C$ to the total
Figure 3.3: Monthly-mean zonal-mean nighttime ice occurrence frequencies based on 8 years of CALIPSO observations, from June 2006 to May 2014, for threshold temperatures of $-40, -50, -60, -70,$ and $-80$°C. Tickmarks are in January. Dashed lines indicate (left to right) the eruptions of Tavurvur in Oct 2006, Kasatochi in Aug 2008, Sarychev in June 2009, and Nabro (13° N) in June 2011.

number of profiles. One might expect that an influx of stratospheric sulfate aerosol into the troposphere may have the strongest impact on the cold and thin cirrus clouds of the uppermost troposphere. Thus, we also provide ice-cloud occurrence frequencies for threshold temperatures of $-50$°C to $-80$°C, as indicated by the colored curves.

In accordance with Sassen et al. (2008), we find that the ice occurrence frequencies exhibit a strong seasonal cycle in all latitude bands. Highest ice abundancies are observed in the Intertropical Convergence Zone (ITCZ), where most convective activity and related transport of water vapor into the upper troposphere occurs. The tropical-mean ice occurrence frequency exhibits a double-peaked shape with maxima in spring and autumn in accordance with the ITCZ equator-crossing times. It also has a weak El Niño signature superimposed: The 2009/10 El Niño event shows up as a pronounced
2009 autumn peak that is right-shifted by a few months towards wintertime. In the midlatitudes, ice is most frequently observed in the respective winter season.

The rightmost dashed line shows the Nabro eruption in June 2011. Also indicated are three other volcanic eruptions with stratospheric SO$_2$ input (see also Figure 3.2). None of the eruptions are followed by abnormally high or low ice occurrence frequencies in any of the five latitude bands. Even thin tropopause cirrus clouds which make up a substantial fraction of the $-70^\circ$C and $-80^\circ$C ice occurrence time series do not show a volcanic signature.

### 3.4.2 Ice–cloud backscatter distributions

Figure 3.4 shows frequency distributions of backscatter measurements of nighttime ice-cloud observations taken over the Northern midlatitude land and ocean areas ($40 - 65^\circ$N) for different seasons based on the CALIPSO L2 CPro dataset. The backscatter measurements shown are the total (perpendicular + parallel) lidar backscatter at 532 nm after attenuation correction. They are provided in the CALIPSO level 2 cloud profile product version 3 provisional (L2 CPro) (Anselmo et al., 2007). The red curves represent frequency distributions of the four seasons following the Nabro eruption, i.e. JJA 2011, SON 2011, DJF 2011/12 and MAM 2012. Seasons of the other years are plotted in black.

The histograms contain only positive backscatter values, even though about 2% of the L2 CPro ice crystal backscatter values are negative. Negative values mostly occur when the corresponding attenuated backscatter values are negative as well, which can be the case in weakly scattering parts of clouds (M. Vaughan, personal communication). We have omitted the negative backscatter values in the logarithmic histograms but counted them in the occurrence frequency plots.

There are months, like May 2013, for which much fewer CALIPSO measurements are available than for others, e.g. due to orbit maintenance. In order to account for this circumstance, we have normalized each histogram by the number of profiles that were sampled to construct it. Thus, a hypothetical doubling in ice occurrence frequency would show as a doubling in height of the corresponding backscatter histogram, because ice would occur twice as often in each profile. So a backscatter histogram shifted upwards (downwards) by a significant amount would indicate an increase (decrease) in the ice occurrence frequency. On the other hand, a significant left or right shift of a particular histogram would hint at the occurrence of larger ice crystals or higher ice crystal number densities. We normalized by the number of profiles, rather than by e.g. the
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Figure 3.4: Backscatter distributions of CALIPSO nighttime ice observations over the Northern midlatitudes $40 - 65^\circ$ N (including land and ocean areas). The red curves are the backscatter distributions of the four seasons following the June 2011 Nabro eruption (JJA 2011–MAM 2012). The black curves indicate the remaining seasons. All distributions have logarithmic bin widths of 0.05, i.e. 20 bins per magnitude. In each subpanel, each row pertains to one of the backscatter distributions, e.g. the row starting with “2011” in the DJF panel pertains to the red curve. The left column states the year, while the middle column shows the number of nighttime in-cloud backscatter measurements between $40 - 65^\circ$ N and below $-40^\circ$ C that went into the distribution of the respective season and year. The right column provides the total number of nighttime CALIPSO profiles available between $40 - 65^\circ$ N for the respective season and year; this includes profiles which contain clouds and profiles without any clouds (clear-sky). For example, if for DJF 2011 there were only 2 orbits, each of which had 550 profiles between $40 - 65^\circ$ N that contained 1800 backscatter measurements from clouds below $-40^\circ$ C at CAD= 100, the DJF 2011 row would read “2011 3600 1100”, and the DJF 2011 distribution (red curve) would contain only 3600 data points.
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Figure 3.5: As Figure 3.4, but for Northern midlatitudes land areas.

As shown in Figures 3.4–3.6, there is significant interannual variability in the heights of the distributions, i.e. in the number of ice-crystal backscatter measurements per backscatter range per profile, in all seasons. The variability is most pronounced in DJF and MAM which is likely induced by the year-to-year variability of midlatitude cyclone frequency and location. The backscatter distributions do not exhibit any systematic upward/downward or left/right shifts in the posteruptive seasons (JJA, SON, DJF 2011 and MAM 2012) following the June 12, 2011 eruption. The posteruptive backscatter distributions (red curves) do not stand out from among other years, except in DJF 2011 over NH midlatitude oceans and in MAM 2012 over the NH midlatitude land areas. Over NH midlatitude oceans, the DJF distributions with the second and third highest peaks pertain to volcanically quiescent periods (DJF 2012 and 2013). Given the substantial interannual variability, only the MAM 2012 backscatter distribution might possibly constitute a significant deviation from the interannual mean. For each of the
backscatter bins, we have calculated the mean and standard deviation based on MAM of years 2007–2011, 2013 and 2014. We found that the MAM 2012 backscatter values in the range of 0.0034 and 0.0169 km\(^{-1}\)sr\(^{-1}\) (or equivalently 10\(^{-2.47}\) and 10\(^{-1.77}\) km\(^{-1}\)sr\(^{-1}\)) lie below the 7-year mean by 3 standard deviations (99.7% confidence level). In the same manner, we performed a statistical test for DJF 2011, in which we found that the DJF 2011 backscatter values do not lie outside of 3 standard deviations from the mean, except for one single backscatter bin (the one centered at 10\(^{-2.0032}\) km\(^{-1}\)sr\(^{-1}\) = 0.0099 km\(^{-1}\)sr\(^{-1}\)) which is just about significant at the 3 sigma level.

However, to make a clear statement about the significance of the DJF 2011 and, in particular, the MAM 2012 backscatter anomalies, we would require measurement time series much longer than eight years in order to allow for a better estimate of the mean and standard deviation of the backscatter per profile in each backscatter range and to test the assumption of normality distribution. Moreover, we would need the deviation to show up also in the posteruptive seasons of JJA 2011 and, in particular, SON 2011, as the strongest potential impact of volcanic aerosol on ice clouds is expected in the first six months after the Nabro eruption. By MAM 2012, i.e. 8.5 to 10.5 months after
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the eruption, the stratospheric aerosol burden has decayed approximately to pre-Nabro values, as can be seen in Figure 3.2.

Other than these, there are no potentially significant deviations in post-eruptive seasons, even at lower threshold temperatures of $-50$ and $-60^\circ\text{C}$ (not shown), i.e. even in ice clouds at greater altitudes. We have also evaluated the ice-cloud backscatter distributions at lower latitudes, namely for the latitude bands $15^\circ\text{N}$–$40^\circ\text{N}$ and $15^\circ\text{S}$–$15^\circ\text{N}$ (over land, oceans, and both), and have found no significant post-eruptive deviations for these lower-latitude bands either. We also examined the distributions of the extinction of the CALIOP beam at 532 nm in clouds at temperatures below $-40^\circ\text{C}$ at $40^\circ\text{N}$–$65^\circ\text{N}$, $15^\circ\text{N}$–$40^\circ\text{N}$ and $15^\circ\text{S}$–$15^\circ\text{N}$ (again over land, oceans, and both) and found no further significant post-eruptive deviations in addition to the ones reported above.

The backscatter distributions of Figures 3.4–3.6 consistently show a first mode that peaks at about $7 \cdot 10^{-3}$ km$^{-1}$ sr$^{-1}$. A second mode is present which peaks at about $3 \cdot 10^{-4}$ km$^{-1}$ sr$^{-1}$ and is particularly pronounced in springtime and in summer. Comparing our backscatter distributions to distributions of ice-crystal extinction derived from simulations of the global aerosol-climate model ECHAM-HAM (Zhang et al., 2012), we found that no second mode is present in the simulated ice clouds, which lead us to examine whether the observed second mode actually stems from ice-crystal backscatter. We noted that the seasonality of the second backscatter mode matches well with the seasonality of upper tropospheric aerosol extinction that is present in particular in springtime but also in summer at NH midlatitudes in the SAGE II extinction dataset. Thomason and Vernier (2013) reported a similar seasonality in aerosol extinction in the SAGE II data. Springtime is a major mineral dust emission time and a substantial fraction of the uplifted dust is transported to the upper troposphere (Wiacek et al., 2010). Mineral dust aerosol and thin ice clouds have similar backscatter and depolarization characteristics, which is why the CALIPSO cloud-aerosol discrimination algorithm does not work fully reliably on dust aerosol plumes (e.g. Chen et al., 2008). Thus, the second (low-backscatter) mode in the distributions of Figures 3.4–3.6 is likely caused by the presence of aerosol particles misclassified as clouds by the level 2 cloud-aerosol discrimination algorithm.

The low-backscatter mode is also evident in the two-dimensional histograms of Figure 3.8 which are normalized frequency distributions of backscatter and residence temperature of ice-cloud measurements made between $45$–$60^\circ\text{N}$ for the post-eruptive seasons (upper row) and the 8-year mean (lower row) as filled contours. The low-backscatter mode of Figure 3.8 indicates that the particles that cause the scattering reside at tem-
Figure 3.7: Zonal mean scattering ratio (aerosol to molecular) based on CALIOP observations over 30–60° N (panel A) and over 0–30° N (panel B).
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Figure 3.8: Histograms of CALIPSO nighttime backscatter measurements and corresponding temperatures from the GEOS-5 model for 45–60° N for the posteruptive seasons JJA 2011 to MAM 2012 (upper row) and 8-year seasonal means (lower row). To allow intercomparison between histograms, each one is normalized by the number of profiles that contributed to it. The contour lines (in white) have been smoothed by a nearest-neighbor running average to improve readability. They are linearly increasing in steps of 0.0001.

Temperatures of about −50° C. Figure 3.7 A) shows the zonal mean aerosol scattering ratio (relative to molecular backscatter) for altitudes of 8–22 km over 30–60° N based on CALIPSO level 1 measurements after cloud-clearing. The seasonality of the aerosol plume in the upper troposphere is evident. The plume is strongest in late spring/early summer. Its residence altitude of about 9–11 km is consistent with the typical residence temperature of about −53° C (Figure 3.8). Figure 3.7 B) shows the zonal mean aerosol scattering ratio over 0–30° N for comparison.

The prominent second mode in SON of Figure 3.5, peaking at about 6 · 10−4 km−1 sr−1, corresponds to the backscatter distribution of fall 2008. The Kasatochi volcano erupted in the Aleutian Island chain at 52° N, 175° W on 8 August 2008. Besides some volcanic ash, its plume contained mostly sulfate aerosol and resided in the UTLS of the NH mid- and high latitudes until November 2008 (Hoffmann et al., 2010; Wang et al.,
2013). It cannot be ruled out that the Kasatochi aerosols have induced the nucleation of ice crystals. However, a substantial part of the peak is likely caused by parts of the plume being misclassified as cirrus cloud by the CALIPSO cloud retrieval algorithms. Visual inspection of vertical feature mask browse images in September and October 2008 supports the latter.

3.4.3 Interannual variability of seasonal ice–cloud occurrence and altitudes

The frequency distributions of Figure 3.8 are histograms of logarithmic bin width 0.05 for backscatter and of 1° C bin width for temperature. Each histogram bin provides the number of nighttime backscatter measurements in the indicated backscatter and temperature ranges, normalized by the number of profiles that contributed to the histogram. As indicated in the Figure, for NH midlatitude ice clouds, temperature–backscatter combinations of between $-40$ to $-50$° C and about $10^{-2}$ km$^{-1}$ sr$^{-1}$, (corresponding to an extinction of about 0.2 km$^{-1}$) are most frequent. The post-eruptive distributions are very similar to the 8-year means. The contour lines indicate the interannual variability (standard deviation) present in the backscatter-temperature distribution entries (lower row) and the post-eruptive deviation from the mean (upper row). None of the post-eruptive backscatter-temperature bins lies outside of three standard deviations of the 8-year mean, so there is no indication of a significant post-eruptive modification of ice-cloud residence altitudes in the NH midlatitudes.

Figure 3.9 compares the post-eruptive deviations in ice occurrence frequencies with the amount of interannual variability in ice occurrence on a 3° × 3° grid. The threshold temperature used was $-40$° C. As indicated by the standard deviations in the lower row, the interannual variability is highest over the tropical Pacific in SON and DJF, which is a signature of the El Niño-Southern oscillation, and lowest in subtropical regions with low ice-cloud occurrence, such as the summertime Southern hemisphere subtropical Indian ocean. The deviations of the post-eruptive seasons (as shown in the middle row) from the 8-year-mean (upper row) have been smoothed by a nearest-neighbors running mean to improve the readability. For each season, the post-eruptive deviations in ice-cloud occurrence generally fall within one standard deviation of the 8-year mean. None of the post-eruptive deviations fall outside of three standard deviations from the 8-year mean in any season.
Chapter 3 Did the 2011 Nabro eruption affect the optical properties of ice clouds?

Figure 3.9: Interannual variability of seasonal nighttime ice occurrence frequencies. Upper row shows mean seasonal ice-cloud occurrence frequencies from eight years of CALIPSO observations. Middle row presents absolute values of deviations from the 8-year seasonal means (shown in the upper row) of the posteruptive seasons JJA 2011 to MAM 2012. Lower row shows seasonal standard deviation calculated from eight years of CALIPSO observations. Regions without data are shaded in gray. The upper colorbar refers to the upper row, the lower colorbar to the middle and lower rows.
Figure 3.10: Seasonal distributions of aerosol backscatter and depolarization of CALIOP nighttime measurements taken between 40–65° N for years 2010–13 at temperatures below −40°C and CAD scores between −70 and −100. The UTLS was largely volcanically unperturbed in all seasons of 2010–13 except for the Nabro post-eruptive period (JJA 2011–MAM 2012). Each backscatter-depolarization histogram is normalized by the highest number of counts occurring in its bins: dark-red histogram pixels correspond to 90–100% of the highest number of counts, while dark-blue pixels correspond to 0–10% of that value. The numbers in the panels indicate season and year, the number of measurements (just below) and the number of profiles that contributed to the histogram.
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There may be instances when CALIOP measurements of clouds, in particular of thin cirrus clouds, can be misclassified as aerosol by the CALIPSO level 2 CAD algorithm. To investigate whether our results are affected in this manner, we have studied the backscatter and depolarization distributions of the CALIOP measurements that were classified as ‘aerosol’ in the CALIPSO level 2 aerosol profile product version 3 provisional (L2 APro). All CALIOP signal returns other than from ‘clear air’ or from the surface are classified as either ‘aerosol’ or ‘cloud’ by the CAD algorithm. ‘Aerosol’ measurements are provided in the L2 APro product, while ‘cloud’ measurements are in the L2 CPro product. Therefore, if significant amounts of thin cirrus clouds were misclassified in a posteruptive season, this would show up in the ‘aerosol’ backscatter-depolarization histograms of that season. Figure 3.10 shows backscatter-depolarization distributions of CALIOP nighttime aerosol measurements taken between 40–65°N for years 2010–13 at temperatures below −40°C and CAD scores between −70 and −100 (i.e. high confidence in the classification as ‘aerosol’). We present this time period because the UTLS was largely volcanically unperturbed in all seasons of 2010–13 except for the Nabro posteruptive period (JJA 2011–MAM 2012), see Figure 3.2. In Figure 3.10, each backscatter-depolarization histogram is normalized by the highest number of counts occurring in its bins. Dark-red histogram pixels correspond to 90–100% of the highest number of counts, while dark-blue pixels correspond to 0–10% of that value. As in Figures 3.4–3.6, the numbers in the panels of Figure 3.10 indicate season and year, the number of measurements (just below) and the number of profiles that contributed to the histogram. Typical lidar depolarization ratios observed for thin cirrus clouds at temperatures below −40°C are 0.35 ± 0.1 (Sassen and Benson, 2001) and typical backscatter values are below $10^{-3}$ km$^{-1}$ sr$^{-1}$. Figure 3.10 shows no changes in the thin-cirrus region thus defined in the posteruptive seasons of JJA 2011–MAM 2012 as compared to the corresponding volcanically quiescent seasons. Rather, JJA 2011 is somewhat shifted towards lower depolarisation ratios because comparatively large amounts of sulfate droplets from the Nabro eruption (which have depolarisation ratios well below 0.1) are detected in that season, so relatively more aerosol measurements at low depolarisation ratios were made in JJA 2011 than in JJAs of other years. In a similar manner, we have investigated the backscatter-depolarization distributions of 15–40°N and 15°S–15°N, and have not found significant amounts of cirrus cloud measurements misclassified as aerosol in those latitude bands either. Moreover, we found no systematic misclassifications of thin cirrus clouds as aerosol in the CALIPSO browse images of the posteruptive seasons.
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3.5 Conclusions

We have analyzed eight years of CALIPSO backscatter and extinction data characterizing ice clouds and find that the June 2011 eruption of the Eritrean Nabro volcano did not cause a statistically significant modification of the optical properties (total backscattering and extinction), occurrence frequencies or residence altitudes of ice clouds on a global scale, even in the uppermost troposphere. We conclude that the eruption had no significant volcanic aerosol-ice cloud radiative effect in the four seasons following it.

According to classical homogeneous nucleation theory, the largest droplets of any sulfate aerosol population are the ones to freeze first (Pruppacher and Klett, 1997; Kärcher and Lohmann, 2002; Cirisan et al., 2013). Simulations of the Nabro sulfate plume performed with the global chemistry-climate model SOCOL-AER (Sheng et al., 2015) indicate that the number densities of the largest sulfate aerosol droplets (≈ 0.1 µm) increased by a factor of 3–4 in the NH midlatitude tropopause region at the lower edge of the plume (and by more than a factor of 10 at higher altitudes) in the months following the Nabro eruption. These increases are consistent with measurements of aerosol number densities made over Laramie (41° N), Wyoming made on 1 June, 28 July and 4 November 2011 (Supporting Online Material of Bourassa et al., 2012). In view of such a large increase in sulfate aerosol abundance in ice-forming air masses, our results suggest that the optical properties of ice and cirrus clouds are at most weakly dependent on the sulfate droplet number density and size. This is in agreement with the analysis of Luo et al. (2002) and with the simulations of Jensen et al. (1994) who found that temperature and cooling rates are the main determinants of ice crystal number density.

Our study does not exclude the possibility of local ice-cloud modifications like those observed by Sassen (1992). CALIPSO’s inherent resolution is 30 meters vertically and about 70 meters horizontally, corresponding to the diameter of the lidar’s ground footprint. The L2 CPro data employed in this study are provided at a resolution of 60 meters vertically and 5–80 km horizontally. In order to avoid sampling artefacts and reduce interannual variability, we have evaluated the L2 CPro data on coarse 3° × 3° grids or as zonal means. While ice-cloud modifications on scales below this resolution cannot be excluded, they would have been locally limited effects.
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Chapter 4

Validation of Ice-Cloud Properties derived from CALIPSO observations in the aerosol-climate model ECHAM6-HAM2

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Abstract

We have compared the ice-cloud occurrence frequencies of five years of ECHAM6-HAM2 simulations to eight years of observations from the satellite-borne elastic backscatter lidar CALIOP. We find that there is generally good agreement between the simulated and the observed ice occurrence frequencies. On the other hand, simulated ice cloud occurrences are clearly underestimated in the tropics and overestimated in parts of the wider Taklamakan region and over Eastern Antarctica. Moreover, the altitudes of the highest ice clouds are overestimated by 1–2 km at most latitudes which may bias the ice-cloud radiative forcing by several W/m².

4.1 Introduction

Ice clouds are an important factor in the Earth’s radiation budget. They scatter and absorb solar radiation, absorb thermal radiation and are emitters in the infrared part of the spectrum. Depending on their altitude and optical thickness, ice clouds can have a radiative warming or cooling effect on surrounding air masses and the surface (e.g., Corti and Peter, 2009; Kübbeler et al., 2014). Several ice-cloud formation mechanisms and morphologies exist. Ice may form when sufficiently moist air is cooled to temperatures at which ice nucleation is possible. Cooling can take place e.g. along a warm front and in warm conveyor belts, during convective transport and in orographic uplifts of air masses. In the context of the present study, ‘ice clouds’ comprise all predominantly ice-containing clouds or parts of clouds, i.e. all cirrus cloud types (such as cirrostratus, cirrocumulus) and glaciated parts of mixed-phase clouds including anvil clouds.

Supercooled liquid water and supercooled aqueous solutions can exist in the atmosphere at temperatures well below the melting point. Homogeneous nucleation is a stochastic process of ice-germ formation in the supercooled liquid in the absence of an insoluble substance that could provide a surface for ice nucleation to take place. The rate of homogeneous ice nucleation can be parameterized as a function of water activity only, with micrometer-size droplets of pure water freezing at about −38°C (Koop et al., 2000). A recent assessment of various homogeneous freezing parameterizations was provided by Ickes et al. (2015).

Heterogeneous nucleation is the process of ice-germ formation at the surface of an insoluble ice nucleus (IN). Typical atmospheric IN types are mineral dust, soot, and bioaerosol such as pollen. The INs provide a surface for the ice germ to form on and
thereby lower the energy barrier that needs to be overcome for nucleation to be possible. Depending on the IN type, ice formation may occur at temperatures as high as about $-2^\circ$ (Hoose and Möhler, 2012). Cziczo et al. (2013) found mineral dust and metallic aerosol particles to be the most efficient IN types. They also suggest that heterogeneous nucleation is the dominant atmospheric freezing mechanism. Besides nucleation, ice-crystal number densities can also be enhanced by splintering: They may substantially exceed IN number densities due to riming-splinting in mixed-phase clouds (Cantrell and Heymsfield, 2005).

The objectives of this study are twofold: First, to validate the properties and abundance of ice clouds as simulated by the cloud-aerosol general circulation model (GCM) ECHAM6-HAM2 (Lohmann and Ferrachat, 2010; Zhang et al., 2012), using an ice-cloud climatology derived from the aerosol-cloud lidar CALIOP. And second, to assess the simulated heterogeneous ice nucleation on mineral dust in the Taklamakan desert and compare observed and simulated ice-cloud occurrences in the wider Taklamakan region.

4.2 Ice-cloud data used

4.2.1 CALIOP data

The Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) elastic backscatter lidar onboard the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) satellite was designed for the study of the optical and microphysical properties of aerosol and cloud particles. It has been collecting backscatter profiles at 532 nm and 1064 nm from particles suspended in the atmosphere since 13 June 2006. CALIOP also measures the depolarization in the 532 nm channel. The lidar is near-nadir pointing and provides measurements at a vertical resolution of 60 m and a horizontal resolution of 5 km. Its ground footprint is about 70 m in cross-section. CALIPSO orbits Earth about 16 times a day at an equatorial altitude of 705 km and with a near-global coverage of up to $82^\circ$ N to $82^\circ$ S. Two adjacent ground tracks are separated by about 100 km in the midlatitudes. CALIPSO has a ground track repeat cycle of 16 days. Its maximal cross-track error is 10 km.

To illustrate the spatial coverage, ground tracks of the nighttime orbits flown by CALIPSO over Southern Asia are shown in Figure 4.1. As the orbit repeat cycle is 16 days, several ground tracks are plotted on top of each other. A $3^\circ \times 3^\circ$ grid box is passed over about four times a month in the midlatitudes. So one $3^\circ \times 3^\circ$ pixel of an eight-year
Figure 4.1: Ground tracks of all nighttime CALIPSO orbits over Southern Asia. The shaded area indicates the location of the Taklamakan desert.

seasonal-mean map represents the mean over the profiles in that grid box of about 96 orbits.

CALIOP has a large linear range that allows backscatter measurements over 5 orders of magnitude in order to detect backscatter from molecules, aerosol and cloud particles (Winker et al., 2007, 2009). The CALIOP backscatter and depolarization measurements have been subjected to the CALIPSO level 2 retrieval algorithm in order to distinguish clear-air, cloud particles and aerosol particles from each other (Young and Vaughan, 2009; Vaughan et al., 2009). We studied eight years of nighttime measurements of particulate backscatter (June 2006 to May 2014) based on the CALIOP level 2 cloud profile product version 3 provisional (L2 CPro) (Anselmo et al., 2007).

Misclassification of aerosol layers as cloud layers and vice versa is an important error source when calculating ice-cloud occurrence frequencies derived from satellite measurements including CALIOP. The cloud-aerosol discrimination (CAD) score introduced by Liu et al. (2009) is a measure of the degree of confidence in the CAD algorithm of Vaughan et al. (2009). We have filtered the backscatter measurements according to the CAD score, leaving only measurements at the highest CAD score confidence levels (CAD score = 100) in the dataset to be analysed. Additionally we make use of tropospheric temperature data from the version 5 Goddard Earth Observing System Model data assimilation system (GEOS-5) of the NASA Global Modeling and Assimilation Office. In order to eliminate polar stratospheric cloud (PSC) pixels from our dataset, we have removed all backscatter measurements above 14 km altitude poleward of 65° N and S. Day-night differences in cloud properties or abundance derived from CALIOP mea-
measurements may reflect actual differences, but could also be a consequence of the lower daytime signal-to-noise ratio. Therefore, we evaluate nighttime measurements only, as they provide the highest signal-to-noise ratios.

Below about $-38^\circ$C, supercooled atmospheric water droplets will freeze homogeneously. We take $T_{\text{hom}} = -40^\circ$C as the homogeneous nucleation threshold temperature in order to allow for possible freezing point depressions in aqueous solution droplets. We define the ice occurrence frequency on a $3^\circ \times 3^\circ$ grid as the number of CALIOP profiles per grid cell that contain one or more in-cloud backscatter measurements at temperatures below $-40^\circ$C, divided by the total number of profiles in that grid cell. The ice occurrence frequency defined in this way is not sensitive to the maximal optical thickness that the lidar can penetrate, because, as long as there is an ice-cloud (no matter how thick), at least its uppermost layer will be detected by the lidar.

4.2.2 Climate-aerosol model ECHAM6-HAM2

The model ECHAM6-HAM2 (Lohmann and Ferrachat, 2010; Zhang et al., 2012) can simulate water, mixed-phase and cirrus clouds, the evolution of aerosol populations and their interactions with the aforementioned cloud types. It contains a two-moment scheme for cloud droplets and ice particles (Lohmann et al., 2008) coupled to a two-moment aerosol scheme (Vignati et al., 2004; Stier et al., 2005) which simulates the evolution of aerosol populations of sulfate, sea-salt, mineral dust, black and organic carbon in terms of aerosol number and mass densities. The mineral dust emissions in ECHAM6-HAM2 are computed dependent on simulated wind speeds and surface moisture based on (Tegen et al., 2002). The ice nucleation scheme simulates the competition between homogeneous nucleation of aqueous solution droplets, immersion freezing of coated dust particles, deposition nucleation on uncoated dust particles and depositional growth on pre-existing ice (Kübbeler et al., 2014). The homogeneous freezing parameterization is based on Koop et al. (2000). The heterogeneous freezing parameterization is derived from Möhler et al. (2006, 2008).

The simulations were performed with model version ECHAM6.1-HAM2.1 at resolution T63L31. The seasonal means were computed based on 5 years of simulations. Climatological monthly-mean sea-surface temperatures, sea-ice, greenhouse gas concentrations and solar forcing were prescribed for the simulations. The ice occurrence frequency was calculated for each vertical column based on the horizontal cloud fractions of the column’s grid boxes at temperatures below $-40^\circ$C. It states how often at least one non-
4.3 Model validation

4.3.1 Ice occurrence frequencies

Figures 4.2 and 4.3 show seasonal ice occurrence frequencies as observed by CALIOP and simulated in ECHAM6-HAM2. There is generally good agreement between the observed and the simulated ice cloud occurrences. The locations and extent of ice occurrence minima in the subtropical dry zones that are characterized by low relative humidity due to the dryness of the descending Hadley circulation air masses are generally well represented. Ice formation is strongest over areas of tropical deep convection, in particular over the Tropical Warm Pool in the Eastern Indian and Western Pacific Oceans, and over tropical South America and Africa. Ice is also dominant in regions where upper tropospheric water vapor is enhanced due to detrainment from tropical convective systems. As expected, the observed ice occurrence frequency distribution of Figure 4.2 matches well with the climatology of upper tropospheric water vapor derived from the Microwave Limb Sounder on the Upper Atmosphere Research Satellite and the International Satellite Cloud Climatology Project (ISCCP) high thick cloud cover climatology, as presented in Figure 5 of Read et al. (1995).
The seasonal shift of the Intertropical Convergence Zone (ITCZ) is evident from the latitudinal shifts of the tropical regions of strongest convective activity and highest ice-cloud occurrence, as can be seen from Figure 4.2. The shift of the ITCZ is well represented in the simulated ice-cloud occurrences. However, the simulated ice-cloud occurrences are substantially too low in the tropical convective regions, i.e. over tropical South America, tropical Africa and the Warm Pool. This may be related to the convective ice formation scheme or to the vertical updraft velocities simulated in tropical convective systems.

The ice-cloud occurrence near the Southern and Western borders of the Taklamakan desert (near 37° N, 82° E, see Figure 4.1) is overestimated in the ECHAM6-HAM2 simulations in all seasons, in particular in DJF and MAM. The ice-cloud occurrence over the Sahara desert is overestimated in MAM. This might be related to the amount of mineral dust emissions or the heterogeneous ice nucleation parameterization.

The L2 CPro product contains backscatter from polar stratospheric clouds which were not fully removed by the level-2 retrieval algorithm. The additional PSC filter employed in this study removes part of these remnants — but not all, as explained below. Therefore, the observed ice occurrence frequencies shown in Figure 4.2 are somewhat overestimated over Antarctica in JJA and SON due to some PSCs that were misclassified.
as ice clouds. Compared to the CALIOP observations, the simulated ice-cloud occurrence is overestimated over parts of Eastern Antarctica in MAM, JJA and SON.

The interannual variability of the seasonal ice-cloud occurrence frequency, as shown in Figure 4.4, is generally underestimated by the simulations shown in Figure 4.5. Rims of high standard deviations shown in the observed variability of Figure 4.4 are likely sampling artefacts, possibly related to the relatively low numbers of nighttime observations available at high latitudes of the respective summer hemisphere. We will clarify this in the further course of this study. Clearly visible is the strong interannual variability in ice-cloud occurrence over the central Pacific in DJF. It is caused by oscillations in the sea-surface temperatures of the equatorial Pacific that are related to the El Niño Southern Oscillation (ENSO) and induce longitudinal shifts of the regions of strongest convection and ice formation. An El Niño signal also seems to be present in the simulations at 15°N–30°S and 120°W–160°E in DJF. It somewhat underestimates the observed interannual ice-cloud variability associated with ENSO and is spatially less confined. The simulations also exhibit strong year-to-year variability in the ice cloud occurrence over the Indian ocean that might be related to simulated monsoon activity. There are further regions of strong interannual variability in simulated ice occurrence, such as over the tropical Atlantic in SON and over Eastern Antarctica in DJF.
Figure 4.5: Seasonal standard deviations of ice-cloud occurrence frequencies from five years simulated in ECHAM6-HAM2.
Figures 4.6–4.7 show observed and simulated zonal-mean ice occurrence frequencies. The model captures the general structure of the ice occurrence distribution, including the maxima observed in the tropics and at higher latitudes, and the minima observed in the subtropics. The seasonal shift of the ITCZ and the associated latitudinal shift of the tropical ice occurrence maxima agree well with the observations. However, the simulations substantially underestimate the maximum ice occurrence frequencies in the tropics. Moreover, the altitude of the simulated tropical maximum lies about 3 km below the observed maximum in all seasons. The simulated zonal-mean ice occurrence distributions exhibit a larger spread in vertical extent, and boundaries that are more smeared out than in the observed distributions. The simulations overestimate the altitudes of the highest cloud tops by about 1–2 km at most latitudes, which might induce a bias of several W/m² to the simulated ice-cloud radiative forcing, see Figure 4 of Corti and Peter (2009).

Not all PSCs observed by CALIPSO over Antarctica in JJA and SON have been removed by the above PSC filtering criteria, as can be seen in Figure 4.6. We will lower the maximum allowable cloud top altitude in the further course of this study to an altitude just above the average local austral wintertime tropopause, e.g. to a maximum 12 km below 65° S, for further analysis. This filtering modification will further reduce the
Figure 4.7: Zonal-mean ice occurrence frequencies over 5 years simulated in ECHAM6-HAM2.
observed ice occurrence frequencies shown over Antarctica in Figure 4.2. The simulations also exhibit ice clouds in the lower Antarctic stratosphere at altitudes as high as 20 km in austral winter and spring time, as shown in Figure 4.7. The cause of this still needs to be investigated.

4.4 Conclusions and Outlook

4.4.1 Conclusions

Using an ice-cloud climatology derived from eight years of measurements taken by the aerosol-cloud lidar CALIOP, we have compared the observed occurrence frequencies of ice clouds to simulations made with the cloud-aerosol GCM ECHAM6-HAM2. The observed and simulated ice occurrence frequencies generally match well. However, the simulated ice cloud occurrence frequencies are clearly underestimated in the tropics and overestimated in the South and in the West of the Taklamakan desert as well as over Eastern Antarctica. We also found that the altitudes of the highest ice-cloud tops are overestimated by 1–2 km at most latitudes. This may bias the ice-cloud radiative forcing by several W/m².

We will analyze further the causes of the mismatches between observed and simulated ice-cloud occurrence frequencies and will also validate the optical properties and residence altitudes of the simulated ice clouds in the further course of this study.

4.4.2 Outlook

We will continue the present study by the following steps:

- **Discarding uncertain measurements.** We will test the sensitivity of the CALIOP ice occurrence frequencies to quality-control screening based on the relative measurement uncertainties, i.e. determine how the CALIOP-derived ice occurrence frequencies change if backscatter measurements with relative uncertainties of greater than, e.g., 100% are disregarded. Figure 4.8 shows the relative uncertainties associated with CALIOP backscatter measurements in ice clouds for an example case (NH midlatitudes in May 2010). The red line indicates measurements with uncertainties of 100%. As can be seen in the Figure, most measurements have relative uncertainties smaller than this value. However, large relative uncertainties can arise at low signal-to-noise ratios, in particular at very low backscatter values,
such as in subvisible cirrus clouds, at cloud edges and in cloud regions that are situated below optically thick cloud regions. In the latter case, the large optical thickness of an upper-level cloud region causes strong attenuation of the lidar beam and thereby weakens the return signal from the cloud region situated below it. Typically, large relative uncertainties arise for measurements at cloud boundaries, in particular the lower cloud edges, as shown in Figure 4.9. The backscatter uncertainty is calculated as explained in Young (2010), starting from the lidar equation and based on the assumption that all uncertainties are random and uncorrelated. When discarding measurements with higher relative uncertainties, one should keep in mind that we might miss out on some very thin ice clouds and, when calculating cloud optical thicknesses, also discard some cloud edges and, therefore, somewhat underestimate the ice-cloud optical thicknesses. Based on Figures 4.8–4.9, we expect the effect on ice occurrence frequencies to be relatively minor.

• **Backscatter and temperature distributions.** We will study the distributions of simulated ice-cloud backscatter and ice-cloud residence temperatures in order
to see how they match with the observed distributions shown in Figures 4.10–4.11. Moreover, simulated zonal-mean ice occurrence frequencies will be compared to the observed ones shown in Figure 4.12.

• **Ice-cloud optical thicknesses.** So far we have been comparing the ECHAM L39 vertical resolution (about 500 m in the upper troposphere) to the finer 60-m resolution of the CALIOP measurements. Naturally, the finer the vertical resolution, the thinner the ice clouds that may be detected, and the greater the number of distinct ice clouds that will be detected. As ECHAM has no subgrid cloud-fraction parameterization in the vertical direction, our current ice-occurrence definition may not be the most appropriate measure for a comparison of ice occurrence simulated at ECHAM6 vertical resolution with CALIOP measurements. This might explain at least part of the substantial underestimation of the simulated tropical ice-cloud occurrences described above. Vertically integrated measures, such as ice-cloud optical thickness, would be less affected by such differences in vertical resolution. They are also more relevant for assessments of the simulated ice-cloud radiative effects. Therefore, we will compare CALIOP ice-cloud optical thicknesses below $-40^\circ$C with the ones simulated in ECHAM6-HAM2 next. It should be kept in

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**Figure 4.9:** Relative uncertainty of backscatter from clouds observed on 16 Oct 2013 at around 15:40 UTC.
mind that, as opposed to the ice occurrence frequency definition we have used so far, vertically integrated measures may be affected by CALIOP’s limit on the maximum penetrable cloud optical thickness.

- **Ice water contents.** Further, we will assess the CALIOP-derived and simulated ice-water content (IWC) below $-40^\circ$C. The CALIOP-derived IWC (in g m$^{-3}$) is related to CALIOP ice-cloud extinction $\varepsilon$ (in km$^{-1}$) by

\[
IWC = 119 \varepsilon^{1.22} \tag{4.1}
\]

according to Heymsfield et al. (2005) who derived this parameterization from in-situ measured IWC and co-located CALIOP extinction measurements.

- **Heterogeneous nucleation on mineral dust.** As shown in Figure 4.1, about nine CALIPSO orbits pass over the Taklamakan desert. We will study the simulated heterogeneous ice nucleation on dust from this region and compare simulations with (a) heterogeneous nucleation only, (b) homogeneous nucleation only, and (c) heterogeneous and homogeneous nucleation. We will assess the simulated ice-cloud occurrences with the ones observed between June 2006–May 2014. The atmospheric circulation will be forced with ERA Interim pressure and winds fields to ensure that the simulated transport of dust is as realistic as possible. Mineral dust emissions in ECHAM6-HAM2 are computed dependent on simulated wind speeds (Stier et al., 2005).
Chapter 4 Validation of CALIPSO-derived Ice-Cloud Properties in ECHAM6-HAM2

Figure 4.10: Backscatter distribution of CALIPSO nighttime measurements in ice clouds observed in the indicated latitude ranges. All distributions have logarithmic bin widths of 0.05, i.e. 20 bins per magnitude. The histograms are normalized by the number of nighttime CALIPSO profiles in the respective season and latitude range. This includes profiles which contain clouds and profiles without any clouds (clear-sky).

Figure 4.11: Histograms of CALIPSO nighttime backscatter measurements and corresponding temperatures from the GEOS-5 model. To allow intercomparison between histograms, each one is normalized by the number of profiles that contributed to it.
Figure 4.12: Monthly-mean zonal-mean nighttime ice occurrence frequencies based on 8 years of CALIPSO observations, from June 2006 to May 2014, for threshold temperatures of $-40$, $-50$, $-60$, $-70$, and $-80$° C. Tickmarks are in June.
Acknowledgments

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Chapter 5

Conclusions and Outlook

5.1 Conclusions

In the present thesis, I have investigated the overall surface climate responses and the responses of ice clouds to sudden increases in the stratospheric aerosol burden caused by major volcanic eruptions. My findings can be summarized as follows:

- The surface temperature and precipitation responses to the eruptions of El Chichón in April 1982 and of Mount Pinatubo in June 1991 have been evaluated as simulated by 17 general circulation models in the framework of the Coupled Model Intercomparison Project phase 5. The objectives of my study were to validate the time lags and coupling strengths of the simulated surface climate responses to changes in the ENSO phase and to validate the simulated post-eruptive temperature and precipitation anomalies over tropical land areas.

Focussing on the responses over tropical land regions, I found that the observed relationship between the ENSO phase, as measured by the Niño 3.4 index, and temperature (or precipitation) can be considered linear to a reasonably good approximation for 1979–2005. The models successfully capture this linearity, though in the case of temperature typically at lower R²s. The observed mean surface air temperature and precipitation increase by 0.16°C and 0.1 mm/day per unit of Niño 3.4 index. Many but not all models simulate temperature and precipitation sensitivities in agreement with this finding.

All models successfully simulate surface air temperature responses delayed by about 4 months relative to the ENSO phase, which agrees well with the observed 4-5 months delay. The strong positive correlation observed between mean temperatures and the ENSO phase (correlation coefficient of 0.75) is generally captured.
well by the models (simulated correlation of 0.71), but there is considerable scatter in the simulated correlation strength across the models (ensemble means of 0.61–0.83). Moreover, the observed precipitation response lags the ENSO phase by 0–1 months. Most of the models appear to simulate a somewhat too fast precipitation response during the El Niño onset (mean simulated lag of -1.8 months), which may be related to a too rapid shift of simulated convective activity towards the ocean. The simulated correlation strength between precipitation and ENSO phase (mean correlation of -0.53) tends to underestimate the observed one (-0.59).

After the Pinatubo eruption, the observed ENSO-removed surface air temperature and precipitation decreased by about 0.35 K and 0.25 mm/day, whereas no significant decrease in either variable was observed after El Chichón. The models generally capture this behavior, even though with large scatter. They appear to somewhat overestimate the precipitation response to El Chichón. The stratospheric AOD increase and associated reduction in evaporation after the El Chichón eruption seem to have not been large enough to result in temperature or precipitation responses beyond the level of natural variability.

- By analyzing eight years of backscatter measurements taken by the satellite lidar CALIOP, I have investigated possible sulfate-aerosol effects on cirrus clouds and glaciated parts of mixed-phase clouds. The focus was on the eruption of the Nabro volcano in June 2011 which released 1–1.5 Mt of sulfur dioxide into the lower stratosphere and was the largest volcanic eruption since Pinatubo in 1991 in that regard.

I found that the Nabro aerosol did not cause a statistically significant modification of the optical properties accessible to CALIOP, neither did the abundance or residence altitudes of ice clouds on a global scale reveal significant changes, not even in the uppermost troposphere. Accordingly, the Nabro eruption induced no significant volcanic aerosol-ice cloud radiative forcing effect in the four seasons following it.

The largest droplets of any sulfate aerosol population are the ones to freeze first. Simulations of the Nabro sulfate aerosol plume performed with the global chemistry-climate model SOCOL-AER indicate enhancements of the number densities of the largest sulfate aerosol droplets ($\approx 0.1 \mu m$) by more than a factor of 3 in the months after the eruption.
In contrast to what one might expect, my results suggest that the optical properties and the abundance of ice clouds depend at most weakly on the sulfate droplet number density and size. This is in agreement with the analyses of Luo et al. (2002) and Jensen et al. (1994) and also with the geoengineering-related simulations of Cirisan et al. (2013), which suggest that temperatures and cooling rates are the main determinants of the ice particle number density in ice clouds.

- CALIOP ice cloud observations have also been used in this thesis for a validation of the occurrence frequencies and residence altitudes of ice clouds as simulated in ECHAM6-HAM2. I found that there is generally a good match between the simulated and the observed ice cloud occurrences. However, the simulations show that the ice cloud abundance is substantially underestimated in the tropics and overestimated in regions south and west of the Taklamakan desert as well as over parts of Antarctica. Further, the altitudes of the highest ice-cloud tops are overestimated by 1–2 km at most latitudes. This may bias the ice-cloud radiative forcing by several W/m².

5.2 Outlook

- Posteruptive surface response (Chapter 2). I have used a linear regression model to separate the surface climate responses to posteruptive increases in stratospheric aerosol burden from the surface climate responses to concurrent El Niño events. Similar approaches, also based on linear regressions between an ENSO index and surface climate variables, have been applied by, e.g., Robock and Mao (1995), Trenberth and Dai (2007), Chen et al. (2008), Joseph and Zeng (2008) and Gu and Adler (2011). By adopting a linear relationship between the ENSO and a surface climate variable, it is assumed that (1) the ENSO phase and strength are not significantly affected by climate responses to volcanic eruptions. This seems well justified in view of the lack of observational evidence for a causal relationship between large volcanic eruptions and El Niño events (e.g. Self et al., 1997; Robock, 2000). A second assumption made is that (2) the ENSO index is related to the surface climate variables by a linear-regression relation over tropical land regions. This assumption appears to be justified in view of the strong correlations between the Niño 3.4 index and surface air temperature or precipitation over tropical land areas. It is beyond the scope of this study to investigate potential non-linear rela-
tionships between ENSO and the surface climate variables. A follow-up study in this direction would be a useful accessory.

- **Posteruptive ice cloud response (Chapter 3).** My results are based on eight years of CALIOP observations which include the largest eruption since Pinatubo in terms of stratospheric sulfur dioxide input. Studies of future major volcanic eruptions based on longer space-lidar observational records are desirable to further investigate the impacts of enhanced upper-tropospheric sulfate aerosol loads on the optical and microphysical properties of ice clouds. Moreover, as CALIOP cannot measure particulate and molecular backscatter in an independent manner, the CALIOP retrievals rely on uncertain assumptions about lidar ratios. High spectral resolution lidars (HSRLs), such as the ATLID lidar that will fly onboard ESA’s EarthCare satellite launched around 2017, can measure aerosol and molecular backscatter independently, which allows them to determine particulate backscatter and extinction independently. This will enable more reliable aerosol-cloud discrimination, making volcanic aerosol-ice cloud studies of future large eruptions based on space-borne HSRL measurements desirable.

Finally, I would like to report on the use of Lagrangian transport modelling to investigate the effects of sulfate aerosol from the Nabro eruption on ice-cloud optical properties. As part of the study presented in Chapter 3, I have identified volcanic aerosol plumes in the lowermost stratosphere in the months after the Nabro eruption and tracked their paths into the upper troposphere. In the CALIOP L2 Aerosol Profile data, I tracked, e.g., a large plume within 0–3 km above the local tropopause over Western Russia on 16 Sept 2011. It travelled eastward and reached the Sea of Okhotsk and Vladivostok on 22 Sept 2011. Forward trajectories started at the plume’s position of 18 Sept over central Russia and Northern Kazakhstan (53°–63° N, 67°–97° E, 195 hPa, 21 UTC) matched the subsequent transport path and time of the plume very well. I have run them forward for 18 days and found that a significant fraction of the trajectories entered the upper troposphere between 28 Sept and 1 Oct 2011. None of the trajectories entered the upper troposphere before 26 Sept 2011. The idea was to evaluate ice cloud properties only in those regions where trajectories had recently entered the upper troposphere, in order to enhance the signal-to-noise ratio in the ice-cloud backscatter distributions (Figure 3.4) by sampling only ice clouds affected by the aerosol.
However, there are issues with this approach. First, CALIOP observations are sparse in space and time. The spatial and temporal distance to the nearest CALIOP observation from the trajectory entrance point is on average approximately 1000 km and 8 days in the midlatitudes. Because of the low orbit repetition frequency, most affected regions were likely not sampled while they experienced an aerosol influx from the stratosphere. On the other hand, the residence time of sulfate aerosol in the upper troposphere is on the order of a few up to 15 days (IPCC, 1999), so ice cloud modifications might still be detectable even if the affected region is overpassed days after the aerosol influx. Moreover, even if trajectories enter the upper troposphere close in space and time to a CALIPSO overpass, they may encounter air masses that are not moist enough to form ice crystals. Therefore, forward trajectories should be run that (1) start from a stratospheric aerosol plume just above the tropopause, ideally within 0–2 km distance from the tropopause (if the plume is at a larger distance, the trajectories will usually take much longer than 10 days until they reach the tropopause, which is not desirable because the trajectory uncertainty increases with time). Secondly, the forward trajectories should (2) enter the upper troposphere in close spatial and temporal vicinity (tolerances to be defined) to a CALIOP ice-cloud observation.

The trajectory approach based on criteria (1) and (2) will be limited by the sparsity of CALIOP observations. And it requires a careful definition of the ice-cloud climatology to be used for comparison and statistical testing. The climatology should ideally be constructed only from ice cloud observations that are unaffected by large influxes of stratospheric sulfate aerosol, be it from the Nabro eruption or from any other sufficiently strong eruption such as that of Tavurvur in 2006 or of the Sarychev volcano in 2009 (Figure 3.2). Moreover, if the CALIOP ice-cloud observations identified based on requirements (1) and (2) are not distributed spatially homogeneously, the climatology should account for this. For example, if 70% of the affected observations were over Northern Africa, and 30% over the Mediterranean and Southern Europe, then the ice-cloud climatology should be an accordingly weighted average of the climatologies of those regions. In view of the above issues, I have decided in favor of a statistical approach rather than a trajectory-based one.

• **ECHAM-HAM validation study (Chapter 4).** An outlook for this study is provided in section 4.4.2.
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