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RESEARCH LETTER

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Key Points:

- Springtime cirrus clouds in the northern hemisphere midlatitudes are affected by volcanic aerosol descending from the stratosphere
- More volcanic aerosols result in cirrus with lower ice content, fewer crystals, and less coverage
- These changes may result in reduced warming from the cirrus clouds

Supporting Information:

Supporting Information may be found in the online version of this article.

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Springtime Stratospheric Volcanic Aerosol Impact on Midlatitude Cirrus Clouds

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Abstract Explosive volcanic eruptions can reach the stratosphere and cause elevated concentrations of sulphate particles for months to years. When these particles descend into the troposphere, they can impact cirrus clouds though to what degree is unknown. In this study, we combine three satellite data sets to investigate the impact of downwelling sulphate aerosol on midlatitude cirrus clouds during springtime. The results show that cirrus clouds in the northern hemisphere (NH) have lower ice water content (IWC), ice crystal number concentrations, and cloud fraction (CF) when the aerosol load in the lowermost stratosphere is elevated by volcanism. These changes are largest for the coldest clouds at the highest altitudes. The cirrus clouds in the southern hemisphere on the other hand show no significant changes with downwelling aerosol levels. The reduction in cirrus IWC and CF in the NH implies that volcanic aerosol can cool the climate through reduced warming from cirrus clouds.

Plain Language Summary Explosive volcanic eruptions can inject the gas sulfur dioxide high up into the atmosphere where the gas forms particles, which can stay airborne for months to years. When the particles descend, they can impact clouds at high altitudes, the so-called cirrus clouds. In this study, we combine three satellite data sets to investigate these impacts from 2008 to 2019. We find that cirrus clouds at midlatitudes in the northern hemisphere (NH) have less ice mass, fewer ice crystals, and smaller coverage when affected by volcanic particles descending during springtime. In the SH, we do not find this effect. The changes in the cirrus clouds in the NH are expected to cool the climate since cirrus clouds normally warm the climate. It has long been known that volcanic particles cool the climate by reflecting solar radiation back to space. In this study, we find that the volcanic eruptions may cause further cooling by their impact on cirrus clouds.

1. Introduction

Explosive volcanic eruptions can have long-term effects on climate by injecting particle-forming SO₂ and ash particles into the stratosphere. The ash and the sulphate particles reflect solar radiation and cool the climate. The ash has a lifetime of a few days to months (Niemeier et al., 2009; Vernier et al., 2016) but the sulphate particles can cool the climate for months or years due to their longer residence time in the stratosphere. Once the particles descend into the troposphere, they can impact cirrus clouds, which could mean a further climate effect. Because cirrus clouds have low optical thickness and high altitude, they generally warm the climate (e.g., Stephens & Webster, 1981). However, their radiative effects are sensitive to small changes in their properties that can strengthen their warming or reverse it to cooling (Cirisan et al., 2013). In this study, we investigate the impact on cirrus clouds from several moderate-sized volcanic eruptions between the years 2008 and 2019.

Cirrus clouds can form through the freezing of supercooled solution droplets via a process called homogeneous freezing. The other formation mechanism, heterogeneous freezing, requires ice nucleating particles (INPs) to initiate the freezing. The balance between these two mechanisms remains unclear and could vary depending on the region (Heymsfield et al., 2017). Cirrus clouds formed from heterogeneous freezing generally have lower ice crystal number concentrations (ICNCs) and larger crystals than those formed from homogeneous freezing (e.g., Krämer et al., 2020). This is because there are fewer INPs than solution droplets, and heterogeneous freezing occurs at lower supersaturations than homogeneous freezing. Thus, if heterogeneous freezing occurs, it will inhibit the formation of more ice crystals through homogeneous freezing (Kärcher & Lohmann, 2003).

Volcanic eruptions reaching the stratosphere take different transport paths depending on the latitude of the eruption and what altitude the eruption reaches. The air transport in the stratosphere is determined by the Brewer-Dobson circulation with ascending air in the tropics that turns poleward and starts to descend at midlatitudes.

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Thus, even aerosols originating from tropical eruptions will descend into the troposphere at midlatitudes or high latitudes. This downwelling varies with season (Škerlak et al., 2014), and the strongest impact from the stratosphere on the upper troposphere (UT) occurs during spring (Martinsson et al., 2017). The mixing of the air from the lowermost stratosphere (LMS) into the UT most often occurs in episodic mesoscale disturbances of the tropopause, such as tropopause folds (Appenzeller & Davies, 1992; Stohl et al., 2003).

Volcanic aerosol impact on cirrus clouds will depend on the freezing regime of the cirrus clouds but may also depend on the type of volcanic aerosol. Ash particles from volcanic eruptions can act as INPs (e.g., Mangan et al., 2017), while the sulphate solution droplets could be homogeneously frozen (Jensen & Toon, 1992), both affecting cirrus clouds. Volcanic sulphate particles could also act as INPs if they are neutralized by ammonia and form solid ammonium sulphate particles, which have been shown to initiate ice at cirrus-relevant conditions (Abbatt et al., 2006). Cirrus clouds formed in situ in the UT are expected to be more sensitive to downwelling stratospheric aerosol than liquid origin cirrus clouds that are fed by air from lower levels in the troposphere (Krämer et al., 2020). These combined features imply that the impact from volcanic aerosol on cirrus clouds is highly uncertain.

Previous satellite investigations of volcanic influence on cirrus clouds have come up with varying results. Two investigations after the eruption of Mount Pinatubo in 1991 found no change in the properties of cirrus clouds (Luo et al., 2002; Wylie & Menzel, 1999), while a third study found an increase in high-level cloudiness (Song et al., 1996). Meyer et al. (2015) investigated the impact on cirrus clouds from the Nabro eruption 2011 but found no significant impact on a global scale. Friberg et al. (2015) on the other hand found lower springtime cirrus cloud reflectance in the NH midlatitudes during years with higher volcanic aerosol loading in the LMS.

The aim of this study is to investigate the impact from downwelling volcanic stratospheric sulphate aerosol on cirrus clouds. More specifically, we aim to study the long-term effects of descending volcanic sulphate aerosol on cirrus clouds rather than the immediate short-term impact of ash from volcanic eruptions. In order to do this, we will use a unique satellite data set of aerosol in the LMS and combine this with two different satellite data sets of cirrus clouds. The investigation will focus on springtime cirrus clouds at midlatitudes as this is the region and season when the upper tropospheric aerosol is dominated by downwelling stratospheric aerosol.

2. Data Sets and Methods

In this section, we describe the aerosol data from the LMS as well as the two cirrus satellite data sets.

2.1. Stratospheric Aerosol Data

The aerosol data are compiled from nighttime Ivl.1b v.4–10 backscattering data from the CALIOP (Cloud-Aerosol Lidar with Orthogonal Polarization) instrument. The data were corrected for attenuation from molecules and particles (Friberg et al., 2018). Ice clouds in the stratosphere were removed using the depolarization ratio (Friberg et al., 2018; Vernier et al., 2009), and data within 2 km of the tropopause were excluded to prevent influence from tropospheric aerosol and hygroscopic growth (Sandvik et al., 2021). The aerosol scattering ratio (total to molecular backscattering) in the remaining LMS (SR_{LMS}) was used to represent the variation in aerosol load in air transported to the UT. SR is suitable since the data are averaged over a height interval that is relative to the tropopause and since the pressure at the midlatitude tropopause varies by more than a factor of two.

2.2. DARDAR Cloud Data

The DARDAR (raDAR/liDAR) data set (Delanoë & Hogan, 2010), combining the CALIOP lidar instrument and the CloudSat radar (Stephens et al., 2002), is used to investigate the microphysical properties of the cirrus clouds. DARDAR provides vertical distributions of cloud properties and in this study, we use the two ice cloud retrieval data sets, such as DARDAR-CLOUD (Delanoë & Hogan, 2010) and DARDAR-NICE (Sourdeval et al., 2018). We utilize the Level 2 products, which have a vertical resolution of 60 m and a horizontal resolution of approximately 1.7 km (Sourdeval et al., 2018). Data from 2008 to 2016 are used though there are missing data during 2011 and 2012 when CloudSat was malfunctioning. After CloudSat became operational again in 2012, it supplied data only during daytime and we therefore only use daytime data in this investigation. Extending this analysis beyond 2016 appeared unreliable due to instrumental issues.

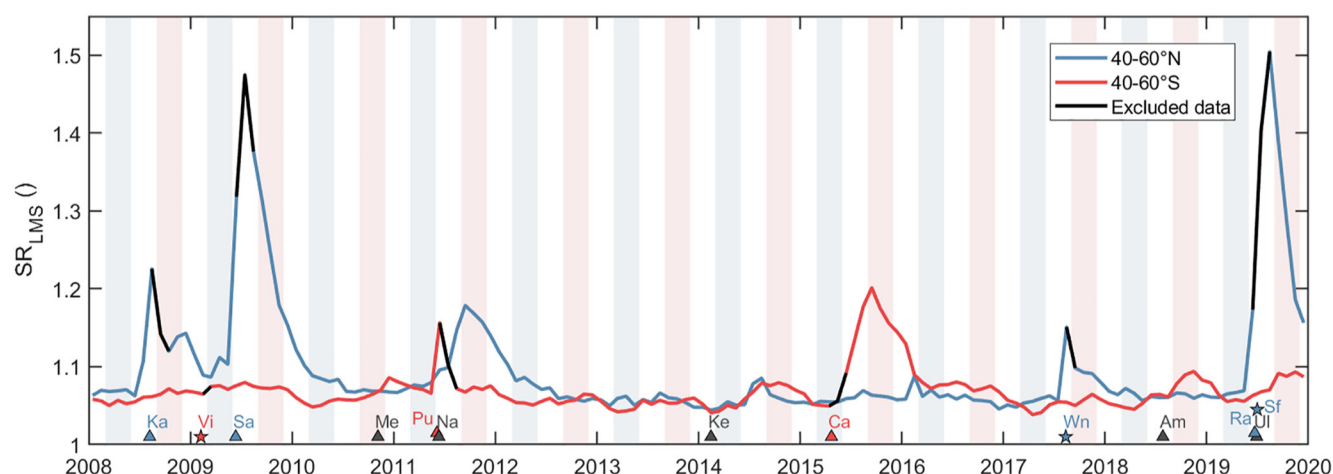


Figure 1. The aerosol scattering ratio in the LMS (SR_{LMS}) at both midlatitude hemispheres. The black parts of the lines show data that have been excluded due to fresh volcanic aerosol at the midlatitudes. The shaded regions show the spring season in the respective hemisphere. Triangles denote volcanic eruptions color-coded according to where the eruption took place (blue - NH midlatitudes, red - SH midlatitudes, and black - tropical eruptions). The stars denote forest fires, color-coded in the same manner. The names, dates, and latitude of the volcanic eruptions/fires are given in Table S1 in Supporting Information S1.

In order to isolate cirrus clouds that could be affected by volcanic aerosol, we applied a cirrus filter (described in the SI) to the Level 2 DARDAR data. From the DARDAR-CLOUD data set, we use the effective radius (r_e) and the IWC. The DARDAR-NICE provides data of the number concentration of ice crystals with diameters larger than 5, 25, and 100 μm . From these data, we calculated the ICNC between 5 and 25 μm as well as between 25 and 100 μm .

2.3. Airs Cloud Data

The Atmospheric Infrared Sounder (AIRS) cloud fraction (CF) data set (Version 7; Tian et al., 2020) used here is a Level 3 monthly mean product in $1^\circ \times 1^\circ$ longitude/latitude grids with 12 vertical standard pressure levels (Team & Teixeira, 2013). The data span from 2008 to 2019 in daily Level 3 data compiled into arithmetic monthly mean values. The AIRS product is divided into data collected in the ascending (day) and descending (night) orbit bins. We use both to calculate weighted zonal means at 40° – 60° for the NH and SH. From the standard pressure levels, we use the CFs in three vertical layers from 300 to 150 hPa. At these levels, we assume the CF to be dominated by cirrus (ice) clouds. Further processing of the cloud data sets is described in Text S2 in Supporting Information S1.

3. Results and Discussion

We will start by presenting how the aerosol load in the LMS has varied over the years included in the investigation before moving on to the results regarding cirrus clouds.

3.1. Volcanic Impact on the Midlatitude Stratosphere

During the years included in this study (2008–2019), a number of volcanic eruptions reached the stratosphere. Though none of these were of the 1991 Mount Pinatubo size, they still had a substantial effect on the stratospheric aerosol load, in particular on the LMS (Friberg et al., 2018). The northern midlatitude LMS was affected by volcanic aerosol to a larger degree than the southern midlatitude LMS (see Figure 1). In particular, the eruptions by Kasatochi (2008) and Sarychev (2009) substantially elevated the SR in the NH LMS during the years 2008–2010, but also the Nabro eruption in 2011 had a substantial impact on the NH. In the SH midlatitudes, there was a substantial impact from the Calbuco eruption in 2015 and Ulawun in 2019.

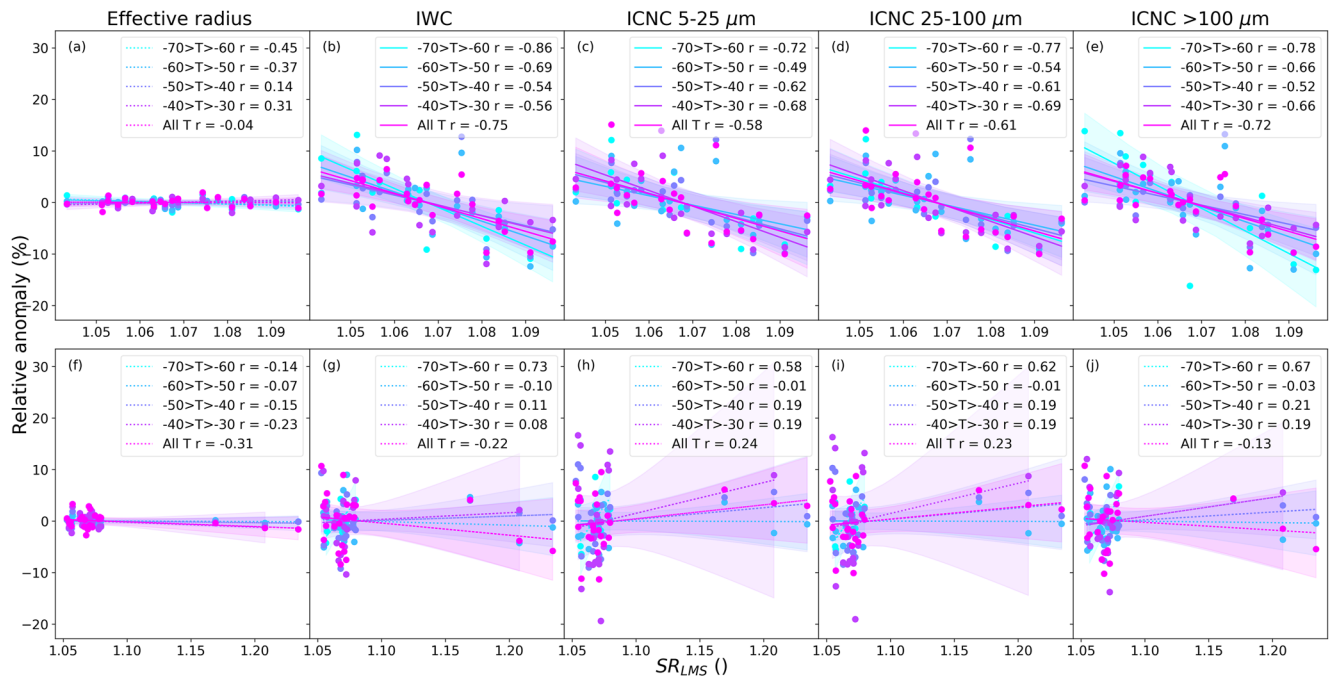


Figure 2. Monthly averaged DARDAR data of microphysical properties of cirrus clouds at different temperatures plotted against the aerosol scattering ratio in the LMS (SR_{LMS}). The DARDAR data are the relative anomaly from a long-term average for each month (see Section Text S2 in Supporting Information S1). The results from the NH (40° – 60°) are shown on the top row and the results from the SH (-60° to -40°) on the bottom row. The lines show linear regressions and solid lines indicate a statistically significant correlation between the two variables (p -value < 0.05) and the shaded areas show the 95% confidence intervals for the predicted values. The correlation coefficients between the variables are shown in the legend. Note the different scales on the x axis for the NH and SH.

3.2. Cirrus Microstructure

In this section, we will look into the volcanic stratospheric impact on midlatitude cirrus clouds microphysics. In order to study the volcanic impact on the cirrus clouds at different temperatures, we divided the DARDAR data into four temperature bins (see Figure 2). This division also implies that the clouds are divided according to height with the coldest clouds residing at the highest altitudes, closest to the downwelling stratospheric aerosol.

The SR_{LMS} has a significant impact on several of the microphysical cirrus properties in the NH. In particular, the IWC decreases when there is a higher aerosol load in the LMS. The relationship is stronger the colder the clouds are, that is, the closer they are to the stratosphere (Figure 2b). The relative change in IWC at the highest/lowest SR_{LMS} values is $\pm 10\%$. The ICNC also decreases with increasing SR_{LMS} at all crystal sizes. The strongest correlations and largest relative changes occur for the largest crystal sizes ($ICNC_{>100}$), which display results similar to those for the IWC. The ICNCs at the smaller sizes have smaller relative changes than $ICNC_{>100}$ but also have the strongest correlations with the SR_{LMS} at the coldest temperatures. The relative changes in r_e with LMS SR are very minor and not statistically significant (Figure 2a). Nevertheless, since the ICNC decreases at all sizes, r_e should not change to a large degree.

Figure 2 shows comparisons of aerosol and cirrus data from the same months of the year. However, downwelling from the higher LMS altitudes may take a few weeks to reach the UT (Friberg et al., 2018). We therefore performed the same analysis with aerosol data shifted to half a month earlier. This decreased the correlations at the highest altitudes somewhat (-0.05 – 0.02), while the correlations at the lower altitudes actually increased slightly (0.02 – 0.03). These results could imply that the subsiding aerosol reaches and impacts the cirrus clouds at lower altitudes somewhat later than those at high altitudes. However, the changes are not statistically significant and the results therefore should be interpreted with caution. Shifting the aerosol data a full month or more only decreased the correlations.

In an attempt to determine if there are regional differences in the effects of the sulphate aerosol on the cirrus clouds, we divided the NH cirrus data into four regions: The Atlantic, Europe-Asia, The Pacific, and America. We use one SR_{LMS} for all regions since volcanic aerosol is expected to be homogeneously distributed over

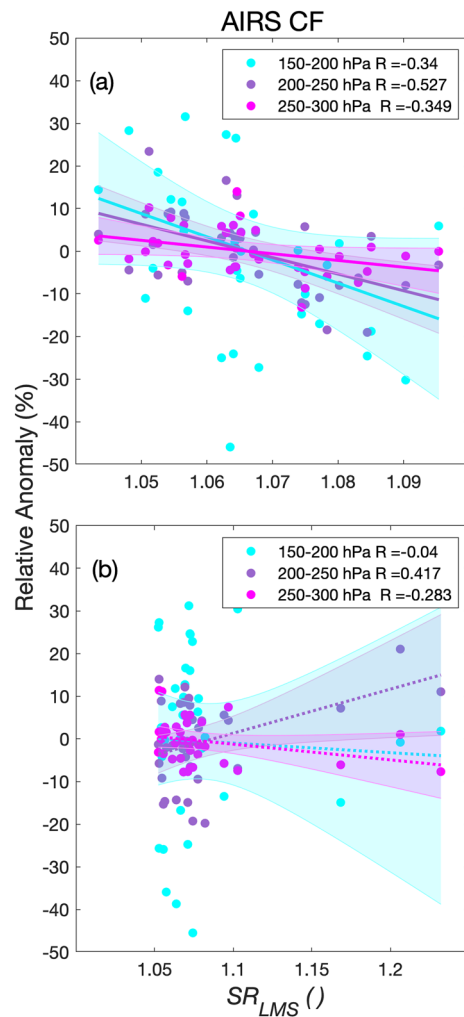


Figure 3. Same as Figure 2 but with the Atmospheric Infrared Sounder (AIRS) cloud fraction at different pressure intervals plotted against the aerosol scattering ratio in the LMS (SR_{LMS}). The AIRS data are the relative anomaly from a long-term average for each month (see Text S2 in Supporting Information S1).

the longitudes. We find that the relationships in Figure 2 are stronger or of similar strength over the Pacific and Atlantic (Figure S4 in Supporting Information S1). The land areas show weaker correlations and over America, the relationships are not statistically significant. These weaker relationships could be explained by a higher frequency of liquid origin cirrus clouds over land, which are expected to be insensitive to downwelling volcanic aerosol. Moreover, the Pacific and Atlantic are the areas most strongly impacted by transport from the LMS to the UT (Škerlak et al., 2014).

In the SH, we do not find statistically significant relationships between any of the cloud microphysical parameters and the SR_{LMS} (Figures 2f–2j). The SH spring data in this study are more often influenced by fresh volcanic aerosol than the NH data set and have a few points with substantially higher LMS aerosol load than the other data points (note the different scales on the x axis between the NH and SH data). This difference between the two hemispheres is caused by the timing of the volcanic eruptions in the NH and SH and where the eruptions occurred (see Figure 1 and Table S1 in Supporting Information S1). The statistics between the cirrus parameters and the SR_{LMS} is highly influenced by the high SR_{LMS} values in the SH, but even when removing these, we find no significant correlation between the microphysical parameters and the SR_{LMS} (see Figures S5 and S6 in Supporting Information S1). These observations indicate that the cirrus cloud microphysics in the SH are less sensitive to volcanic aerosol than those in the NH.

3.3. Cirrus Cloud Fraction

Data from the AIRS satellite sensor are used to determine if the cirrus CF is impacted by the aerosol load in the LMS. In the NH, there is a significant decrease in CF with increasing SR_{LMS} at all pressure levels (see Figure 3). The correlation is not as strong as for the IWC and $ICNC_{>100}$ data but the relative change is larger or similar for the AIRS data. The correlations in the AIRS data set do not increase with altitude, as they were for some of the microphysical parameters. The weaker correlations for the CFs than the microphysical parameters are to some extent expected since the cloud fraction is strongly impacted by the synoptic weather conditions, which are highly variable from year to year. Investigating the CF for different regions did not reveal any stronger CF changes with SR_{LMS} than using all longitudes. As for the DARDAR data, the AIRS trends in the SH are heavily dominated by a few points with high volcanic impact and have weak statistically insignificant correlations.

3.4. Comparison With Previous Studies

The results in this study agree with two previous studies of moderate-sized volcanic eruptions' impact on cirrus clouds. Friberg et al. (2015) found decreasing cirrus cloud reflectance with increasing sulphate aerosol in the LMS. The lower IWC and CF with higher SR_{LMS} found in this study would indeed result in a lower cirrus reflectance. Moreover, Meyer et al. (2015) found lower backscattering from cirrus clouds in the NH during spring 2012, the year after the Nabro eruption. Our results differ from three satellite studies of the 1991 Mount Pinatubo eruption (Luo et al., 2002; Song et al., 1996; Wylie & Menzel, 1999). This could be explained by the larger size of the Mount Pinatubo Eruption and perhaps also by more limited satellite data sets available at that time.

Most modeling studies of volcanic sulfuric acid particles on cirrus clouds do not agree with our results. A study covering many of the eruptions included in this study found an increase in ICNC and a decrease in their size in response to additional sulfur in the LMS/UT (Schmidt et al., 2018). Similarly, Lohmann (2003) found an increase in ICNC in homogeneously formed cirrus clouds in a response to additional sulphate aerosol from the 1991 Mt

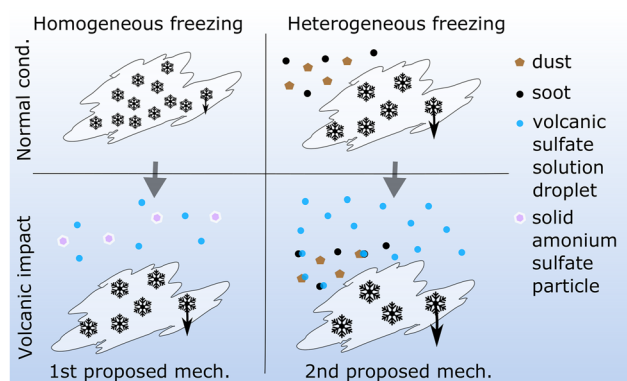


Figure 4. Cirrus clouds under normal conditions formed through homogeneous and heterogeneous freezing are shown in the upper panels. The lower panels show the two different proposed mechanisms of how volcanic aerosol impacts cirrus clouds. Both mechanisms result in larger ice crystals in the cirrus cloud and increased settling velocity (black arrows).

Pinatubo eruption. Jensen and Toon (1992) on the other hand found that the changes in ICNC depend on whether volcanic sulphate aerosols nucleate ice homogeneously or heterogeneously.

3.5. Possible Causes and Implications

The results presented in the Results section point to a hemispheric difference in volcanic impact on cirrus clouds. There are quite a few differences between the hemispheres that are relevant for cirrus clouds. There are more land areas and greater sources of INPs in the NH, in particular because there are more sources of dust (e.g., Storelvmo & Herger, 2014). There is also more air traffic in the NH, which means more emissions of soot at high altitudes (Zhou & Penner, 2014). Soot particles, in particular from air traffic, are considered to be possible INPs (Kärcher et al., 2007). The greater availability of INPs in the NH also means that heterogeneous freezing is likely more common in the NH. Yet another difference between the hemispheres is the larger amount of anthropogenic emissions in the NH (Clarisse et al., 2009).

In the NH, IWC and ICNC decrease with higher SR_{LMS} . We can also see that this is associated with a decrease in cirrus cloud fraction, but these relations are not as strong as the ones seen for the microphysical parameters. The decreases in IWC and CF indicate that either there is less ice formed in the cirrus clouds under volcanic influence or the crystals formed are larger and sediment faster, leading to a smaller amount of ice in the clouds. The first of these explanations would most likely be associated with a change in the atmospheric dynamics impacting temperature or relative humidity. It is unlikely that such changes would act differently in the two hemispheres and we therefore focus on the second hypothesis, that is, microphysical changes of the cirrus clouds.

We have considered several mechanisms in which descending volcanic sulfate could cause the ice crystals in the cirrus clouds to become fewer and larger, thus sedimenting faster. The formation of larger crystals would inhibit the formation of smaller crystals, leading to decreased ICNC also at these sizes (Figures 2c and 2d). This results in a decrease in ICNC at all sizes, leading to very minor changes in r_e (Figure 2a). We will present two explanations for the microphysical changes that we find most plausible. It is still unknown which freezing mechanism that dominates cirrus cloud formation in the NH, and it could vary with altitude and region. One of our explanations is valid in a homogeneous freezing regime and the other is valid in a heterogeneous freezing regime.

The first mechanism assumes that the cirrus freezing regime is homogeneous, that is, the cirrus clouds form through freezing of solution droplets and have many small crystals (see Figure 4). The downwelling volcanic aerosol consists of sulfuric acid and water but may react with trace gases present in the UT, such as ammonia. Ammonium sulfate particles have in laboratory studies been found to act as INPs at temperatures below -38°C and at an ice saturation ratio (S_{ice}) as low as 1.10–1.20 (Abbatt et al., 2006; Baustian et al., 2010; Hoose & Möhler, 2012) well below that of homogeneous freezing (1.40). It has also recently been reported that ammonium nitrate particles can crystallize at $S_{ice} < 1.40$ at cirrus temperatures if some ammonium sulfate is added to the particles (Wagner et al., 2020). Ammonium nitrate has been observed in the UT in the NH (Höpfner et al., 2019). It is thus possible that some of the downwelling volcanic aerosols react with compounds in the NH UT and form INPs. If enough INPs are formed, the freezing regime will switch from homogeneous to heterogeneous, resulting in cirrus clouds with larger but fewer ice crystals (see Figure 4). These will sediment faster, reducing the IWC, ICNC, and CF, as is seen in the NH in this study. The reason why this change would occur only in the NH is the higher emissions of ammonia in the NH compared to the SH.

The second possible explanation mechanism that we propose assumes that the dominant freezing regime in the affected cirrus clouds is heterogeneous with freezing being initiated by INPs, such as dust or soot (see Figure 4). The downwelling sulphate aerosol could coat some of these INPs, making them less efficient as ice nuclei (e.g., Hoose & Möhler, 2012). This would result in fewer INPs present during cirrus formation, which would lead to fewer, larger ice crystals in the cirrus clouds. Such crystals would sediment faster and support the results found in our study. This would not occur in the SH since there are less INPs available and the freezing regime would generally not be heterogeneous.

Which of these explanation mechanisms that are most plausible depends on which freezing regime that is dominant in the affected cirrus clouds in the NH. Aircraft measurements support that heterogeneous nucleation is dominant in some parts of the NH (Cziczo et al., 2013; Gayet, 2004). However, the clouds found to be most strongly affected by downwelling aerosol in this study are the ones at the coldest temperatures and highest altitudes. The INP concentrations should decrease with altitude and it may seem unlikely to find the strongest impact on the cirrus clouds where the INP concentrations are the lowest. It may be that at such high altitudes, homogeneous freezing is the dominant cirrus formation mechanism. Nevertheless, if there are INPs present at the highest altitudes, then the chance of coating these is highest here. There are currently not enough data on which freezing mechanism that is dominant for cirrus clouds at these altitudes in the NH and we therefore do not propose *one* explanation mechanism that we find more probable than the other.

Another possible cause for the contrasting results between hemispheres could be differences in the transport from the LMS to the UT. This transport displays larger seasonal and spatial variation in the NH (Škerlak et al., 2014) than in the SH. The greater variability in the NH transport implies that the cirrus clouds in the NH are exposed to high concentrations of volcanic sulphate aerosol from the LMS during certain seasons and in specific regions. The impact in the SH on the other hand could be more evenly spread both seasonally and regionally. High concentrations of volcanic sulphate aerosol at specific locations could explain the hemispheric difference in the results but also support the proposed microphysical explanations above.

High thin cirrus clouds have a net warming effect on the climate, and a lowering of their IWC and CF (as found in this study) is therefore expected to reduce their warming (e.g., Lohmann & Gasparini, 2017). Thus, volcanic aerosols seem to be able to not only cool the climate by scattering solar radiation, but also through the impact on cirrus clouds at least in the NH. That the volcanic effect on cirrus clouds is confined to the NH means that there may be an asymmetric cooling from tropical volcanic eruptions. An interesting aspect of the two explanation mechanisms presented above is that both contain anthropogenic impact on the atmosphere, through emissions of ammonia and soot particles. Hence, human influence on the atmosphere may have enabled an enhanced volcanic cooling of the climate.

4. Conclusions

Using three different satellite data sets, we found that cirrus cloud properties in the NH are affected by volcanic aerosol downwelling from the stratosphere during spring. The cirrus clouds in the NH have lower IWC, ICNC, and CF during years with higher volcanic impact on the LMS, in particular the clouds at the highest altitudes. The clouds in the SH did not show statistically significant changes with LMS aerosol load. We propose that the changes seen in the NH result from the cirrus clouds having larger ice crystals, which sediment faster, causing a decrease in IWC, ICNC, and CF.

We identify two viable mechanisms that can explain our observations. (a) The formation of larger crystals could be a result of the volcanic aerosol forming IN, seeding cirrus clouds formed through homogeneous freezing or (b) the downwelling volcanic aerosol could coat existing INPs in the UT, providing less INPs for cirrus clouds formed through heterogeneous freezing.

The impact on the climate from a decrease in cirrus IWC and CF is a cooling, indicating that volcanic aerosols do not only cool the climate through scattering of solar radiation but also through the impact on cirrus clouds. These results add to the uncertainty in the present modeling of volcanic climate impact as most models currently include only the direct climate impact of volcanism. The results in this study are also highly relevant in the considerations of deliberate manipulation of Earth's climate through the addition of sulphate aerosol to the stratosphere.

Data Availability Statement

The DARDAR data were provided by ICARE data and service center. The AIRS data were provided by Goddard Earth Sciences Data and Information Services Center (GES DISC) at <https://airs.jpl.nasa.gov/>. The DARDAR cirrus cloud product that we developed in this investigation and the SR_{LMS} data are available at <https://doi.org/10.5878/xdwz-3g72>.

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References

- Abbatt, J. P. D., Benz, S., Cziczo, D., Kanji, Z., Lohman, U., & Möhler, O. (2006). Solid ammonium sulfate aerosols as ice nuclei: A pathway for cirrus cloud formation. *Science*, 313(5794), 1770–1773. <https://doi.org/10.1126/science.1129726>
- Appenzeller, C., & Davies, H. C. (1992). Structure of stratospheric intrusions into the troposphere. *Nature*, 358(6387), 570–572. <https://doi.org/10.1038/358570a0>
- Baustian, K. J., Wise, M. E., & Tolbert, M. A. (2010). Depositional ice nucleation on solid ammonium sulfate and glutaric acid particles. *Atmospheric Chemistry and Physics*, 11, 2307–2317. <https://doi.org/10.5194/acp-10-2307-2010>
- Crisan, A., Spichtinger, P., Luo, B. P., Weisenstein, D. K., Wernli, H., Lohmann, U., & Peter, T. (2013). Microphysical and radiative changes in cirrus clouds by geoengineering the stratosphere: Geoengineering effect on cirrus clouds. *Journal of Geophysical Research: Atmospheres*, 118(10), 4533–4548. <https://doi.org/10.1002/jgrd.50388>
- Clarisse, L., Clerbaux, C., Dentener, F., Hurtmans, D., & Coheur, P.-F. (2009). Global ammonia distribution derived from infrared satellite observations. *Nature Geoscience*, 2(7), 479–483. <https://doi.org/10.1038/ngeo551>
- Cziczo, D. J., Froyd, K. D., Hoose, C., Jensen, E. J., Diaio, M., Zondlo, M. A., et al. (2013). Clarifying the dominant sources and mechanisms of cirrus cloud formation. *Science*, 340(6138), 1320–1324. <https://doi.org/10.1126/science.1234145>
- Delanoë, J., & Hogan, R. J. (2010). Combined CloudSat-CALIPSO-MODIS retrievals of the properties of ice clouds. *Journal of Geophysical Research*, 115, D00H29. <https://doi.org/10.1029/2009JD012346>
- Friberg, J., Martinsson, B. G., Andersson, S. M., & Sandvik, O. S. (2018). Volcanic impact on the climate—The stratospheric aerosol load in the period 2006–2015. *Atmospheric Chemistry and Physics*, 18(15), 11149–11169. <https://doi.org/10.5194/acp-18-11149-2018>
- Friberg, J., Martinsson, B. G., Sporre, M. K., Andersson, S. M., Brenninkmeijer, C. A. M., Hermann, M., et al. (2015). Influence of volcanic eruptions on midlatitude upper tropospheric aerosol and consequences for cirrus clouds. *Earth and Space Science*, 2(7), 285–300. <https://doi.org/10.1002/2015EA000110>
- Gayet, J.-F., Ovarlez, J., Shcherbakov, V., Ström, J., Schumann, U., Minikin, A., et al. (2004). Cirrus cloud microphysical and optical properties at southern and northern midlatitudes during the INCA experiment. *Journal of Geophysical Research*, 109(D20), D20206. <https://doi.org/10.1029/2004JD004803>
- Heysfield, A. J., Krämer, M., Luebke, A., Brown, P., Cziczo, D. J., Franklin, C., et al. (2017). Cirrus clouds. *Meteorological Monographs*, 58, 2.1–2.26. <https://doi.org/10.1175/AMSMONOGRAPH5-D-16-0010.1>
- Hoose, C., & Möhler, O. (2012). Heterogeneous ice nucleation on atmospheric aerosols: A review of results from laboratory experiments. *Atmospheric Chemistry and Physics*, 12(20), 9817–9854. <https://doi.org/10.5194/acp-12-9817-2012>
- Höpfner, M., Ungermann, J., Borrmann, S., Wagner, R., Spang, R., Riese, M., et al. (2019). Ammonium nitrate particles formed in upper troposphere from ground ammonia sources during Asian monsoons. *Nature Geoscience*, 12(8), 608–612. <https://doi.org/10.1038/s41561-019-0385-8>
- Jensen, E. J., & Toon, O. B. (1992). The potential effects of volcanic aerosols on cirrus cloud microphysics. *Geophysical Research Letters*, 19(17), 1759–1762. <https://doi.org/10.1029/92GL01936>
- Kärcher, B., & Lohmann, U. (2003). A parameterization of cirrus cloud formation: Heterogeneous freezing. *Journal of Geophysical Research*, 108(D14), 4402. <https://doi.org/10.1029/2002JD003220>
- Kärcher, B., Möhler, O., DeMott, P. J., Pechtl, S., & Yu, F. (2007). Insights into the role of soot aerosols in cirrus cloud formation. *Atmospheric Chemistry and Physics*, 7(16), 4203–4227. <https://doi.org/10.5194/acp-7-4203-2007>
- Krämer, M., Rolf, C., Spelten, N., Afchine, A., Fahey, D., Jensen, E., et al. (2020). A microphysics guide to cirrus—Part 2: Climatologies of clouds and humidity from observations. *Atmospheric Chemistry and Physics*, 20(21), 12569–12608. <https://doi.org/10.5194/acp-20-12569-2020>
- Lohmann, U. (2003). Impact of the Mount Pinatubo eruption on cirrus clouds formed by homogeneous freezing in the ECHAM4 GCM. *Journal of Geophysical Research*, 108(D18), 4568. <https://doi.org/10.1029/2002JD003185>
- Lohmann, U., & Gasparini, B. (2017). A cirrus cloud climate dial? *Science*, 357(6348), 248–249. <https://doi.org/10.1126/science.aan3325>
- Luo, Z., Rossow, W. B., Inoue, T., & Stubenrauch, C. J. (2002). Did the eruption of the Mt. Pinatubo volcano affect cirrus properties? *Journal of Climate*, 15, 2806–2820. [https://doi.org/10.1175/1520-0442\(2002\)015<2806:dteotm>2.0.co;2](https://doi.org/10.1175/1520-0442(2002)015<2806:dteotm>2.0.co;2)
- Mangan, T. P., Atkinson, J. D., Neuberg, J. W., O'Sullivan, D., Wilson, T. W., Whale, T. F., et al. (2017). Heterogeneous ice nucleation by soufrière hills volcanic ash immersed in water droplets. *PLoS One*, 12(1), e0169720. <https://doi.org/10.1371/journal.pone.0169720>
- Martinsson, B. G., Friberg, J., Sandvik, O. S., Hermann, M., van Velthoven, P. F. J., & Zahn, A. (2017). Particulate sulfur in the upper troposphere and lowermost stratosphere—Sources and climate forcing. *Atmospheric Chemistry and Physics*, 17(18), 10937–10953. <https://doi.org/10.5194/acp-17-10937-2017>
- Meyer, A., Vernier, J., Luo, B., Lohmann, U., & Peter, T. (2015). Did the 2011 Nabro eruption affect the optical properties of ice clouds? *Journal of Geophysical Research: Atmospheres*, 120(18), 9500–9513. <https://doi.org/10.1002/2015JD023326>
- Niemeier, U., Timmreck, C., Graf, H.-F., Kinne, S., Rast, S., & Self, S. (2009). Initial fate of fine ash and sulfur from large volcanic eruptions. *Atmospheric Chemistry and Physics*, 9(22), 9043–9057. <https://doi.org/10.5194/acp-9-9043-2009>
- Sandvik, O. S., Friberg, J., Sporre, M. K., & Martinsson, B. G. (2021). Methodology to obtain highly resolved SO₂ vertical profiles for representation of volcanic emissions in climate models (preprint). *Aerosols/Remote Sensing/Data Processing and Information Retrieval*. <https://doi.org/10.5194/amt-2021-94>
- Schmidt, A., Mills, M. J., Ghan, S., Gregory, J. M., Allan, R. P., Andrews, T., et al. (2018). Volcanic radiative forcing from 1979 to 2015. *Journal of Geophysical Research: Atmospheres*, 123(22), 12491–12508. <https://doi.org/10.1029/2018JD028776>
- Škerlak, B., Sprenger, M., & Wernli, H. (2014). A global climatology of stratosphere–troposphere exchange using the ERA-Interim data set from 1979 to 2011. *Atmospheric Chemistry and Physics*, 14(2), 913–937. <https://doi.org/10.5194/acp-14-913-2014>
- Song, N., Starr, D. O., Wuebbles, D. J., Williams, A., & Larson, S. M. (1996). Volcanic aerosols and interannual variation of high clouds. *Geophysical Research Letters*, 23(19), 2657–2660. <https://doi.org/10.1029/96GL02372>
- Sourdeval, O., Gryspeerdt, E., Krämer, M., Goren, T., Delanoë, J., Afchine, A., et al. (2018). Ice crystal number concentration estimates from lidar–radar satellite remote sensing—Part 1: Method and evaluation. *Atmospheric Chemistry and Physics*, 18(19), 14327–14350. <https://doi.org/10.5194/acp-18-14327-2018>
- Stephens, G. L., Vane, D. G., Boain, R. J., Mace, G. G., Sassen, K., Wang, Z., et al. (2002). The cloudsat mission and the a-train. *Bulletin of the American Meteorological Society*, 83(12), 1771–1790. (Place: Boston MA, USA Publisher: American Meteorological Society). <https://doi.org/10.1175/BAMS-83-12-1771>
- Stephens, G. L., & Webster, P. J. (1981). Clouds and climate: Sensitivity of simple systems. *Journal of the Atmospheric Sciences*, 38(2), 235–247. (Place: Boston MA, USA Publisher: American Meteorological Society). [https://doi.org/10.1175/1520-0469\(1981\)038<0235:cacos>2.0.co;2](https://doi.org/10.1175/1520-0469(1981)038<0235:cacos>2.0.co;2)
- Stohl, A., Wernli, H., James, P., Bourqui, M., Forster, C., Liniger, M. A., et al. (2003). A new perspective of stratosphere–troposphere exchange. *Bulletin of the American Meteorological Society*, 84(11), 1565–1574. <https://doi.org/10.1175/BAMS-84-11-1565>

- Storelvmo, T., & Herger, N. (2014). Cirrus cloud susceptibility to the injection of ice nuclei in the upper troposphere. *Journal of Geophysical Research: Atmospheres*, 119(5), 2375–2389. <https://doi.org/10.1002/2013JD020816>
- Team, A. S., & Teixeira, J. (2013). Aqua AIRS level 3 standard monthly product using AIRS IR-only V6. *NASA Goddard Earth Sciences Data and Information Services Center*. (Type: dataset). <https://doi.org/10.5067/AQUA/AIRS/DATA321>
- Tian, B., Manning, E., Roman, J., Thrastarson, H., Fetzer, E. J., & Monarrez, Z. (2020). *AIRS version 7 level 3 product user guide*. Jet Propulsion Laboratory California Institute of Technology Pasadena. Retrieved from https://docserver.gesdisc.eosdis.nasa.gov/public/project/AIRS/V7_L3_User_Guide.pdf
- Vernier, J., Fairlie, T. D., Deshler, T., Natarajan, M., Knepp, T., Foster, K., et al. (2016). In situ and space-based observations of the Kelud volcanic plume: The persistence of ash in the lower stratosphere. *Journal of Geophysical Research: Atmospheres*, 121(18), 11104–11118. <https://doi.org/10.1002/2016JD025344>
- Vernier, J. P., Pommereau, J. P., Garnier, A., Pelon, J., Larsen, N., Nielsen, J., et al. (2009). Tropical stratospheric aerosol layer from CALIPSO lidar observations. *Journal of Geophysical Research*, 114(D4), D00H10. <https://doi.org/10.1029/2009JD011946>
- Wagner, R., Bertozzi, B., Höpfner, M., Höhler, K., Möhler, O., Saathoff, H., & Leisner, T. (2020). Solid ammonium nitrate aerosols as efficient ice nucleating particles at cirrus temperatures. *Journal of Geophysical Research: Atmospheres*, 125(8), e2019JD032248. <https://doi.org/10.1029/2019JD032248>
- Wylie, D. P., & Menzel, W. P. (1999). Eight years of high cloud statistics using HIRS. *Journal of Climate*, 12(1), 170–184. (Place: Boston MA, USA Publisher: American Meteorological Society). <https://doi.org/10.1175/1520-0442-12.1.170>
- Zhou, C., & Penner, J. E. (2014). Aircraft soot indirect effect on large-scale cirrus clouds: Is the indirect forcing by aircraft soot positive or negative?: Aircraft soot indirect effect. *Journal of Geophysical Research: Atmospheres*, 119(19), 11303–11320. <https://doi.org/10.1002/2014JD021914>