Comparing the Radiative Forcings of the Anthropogenic Aerosol Emissions From Chile and Mexico

Tuuli Miinalainen1 ©, Harri Kokkola2, Kari E. J. Lehtinen1,2, and Thomas Kühn1,2

1Department of Applied Physics, University of Eastern Finland (UEF), Kuopio, Finland, 2Atmospheric Research Centre of Eastern Finland, Finnish Meteorological Institute (FMI), Kuopio, Finland

Abstract

There has been growing interest in the potential of short-lived climate forcer (SLCF) mitigation to reduce near-term global warming. Black carbon (BC), organic carbon (OC), and sulfur dioxide (SO2) are SLCFs which change the Earth’s radiative balance directly by affecting radiation, and indirectly by altering cloud properties. We used the ECHAM-HAMMOZ aerosol-climate model to study the radiative forcings due to mitigating the anthropogenic emissions of BC, OC, and SO2 from Chile and Mexico. Limiting our analysis to areas where these emissions had notable effects on both aerosol and clouds, we found that the total radiative forcings of anthropogenic aerosol emissions are different for Chile and Mexico. This was explained by differences in aerosol emissions, orography, and meteorology in these two countries. Especially the radiative forcing for Chilean emissions was influenced by the persistent stratocumulus cloud deck west of Chile. To reduce the uncertainty of our radiative forcing calculations, we nudged the wind and surface pressure toward pre-generated fields. As nudging affects the calculated effective radiative forcing (ERF), we here used the identifier ERFNDG. Our results indicate that the removal of OC and SO2 emissions caused a positive ERFNDG while the removal of BC emissions caused a positive ERFNDG for Chile, but a negative ERFNDG for Mexico. When accounting for co-emission of other aerosol compounds, reducing BC emissions led to positive ERFNDG in both countries. Compared to China, the removal of anthropogenic SO2 emissions in Chile and Mexico caused a much larger global average ERFNDG per emitted unit mass of SO2.

1. Introduction

Short-lived climate forcers (SLCFs) are compounds that originate either from natural sources or human activity. In broad terms, SLCFs are atmospheric compounds that can have a substantial effect on the climate and global warming, but have a relatively short atmospheric lifetime of a few days to a decade compared to long-lived greenhouse gases (e.g., carbon dioxide [CO2]) which can have lifetimes of hundreds of years. SLCFs include both gaseous compounds such as methane (CH4) and hydrofluorocarbons, and aerosols such as black carbon (BC), organic carbon (OC), and sulfate (Stohl et al., 2015; UNEP, 2011).

Out of all SLCFs, especially BC and CH4 are attributed a warming effect on the climate, as they absorb shortwave (SW) and longwave (LW) radiation, respectively (Smith & Mizrahi, 2013). In addition, many SLCFs, including BC, OC, and sulfate, contribute to air pollution, which has become a central issue in most of the metropolitan areas worldwide (Krzyzanowski et al., 2014). Recently, Burnett et al. (2018) suggested that outdoor particulate air pollution could be attributable to 8.9 million deaths globally in 2015, whereas some previous estimates lie between 2 and 4 million annually (Forouzanfar et al., 2016; Silva et al., 2013). Consequently, SLCF mitigation is seen as an attractive option to improve local air quality, and mitigation of particularly warming SLCF is seen as an option to “buy time” for adapting to global warming (Bowmer et al., 2013; CCAC & UNEP, 2016).

BC is a SLCF that is assumed to play a major role in global warming (AMAP, 2015; Bond et al., 2013). However, many recent studies found that the warming effect of BC aerosol might be lower than originally estimated (Andrews et al., 2017; Lund et al., 2018; Peng et al., 2016; Wang et al., 2016, 2018; Zanatta et al., 2016). BC is emitted during incomplete combustion processes, for example from residential heating with biomass and from vehicle engines. Natural processes, for example, forest fires and volcanic eruptions, release substantial amounts of BC into the air as well (Bond et al., 2013). As a strongly light-absorbing substance, BC alters the Earth’s radiation budget by absorbing solar radiation and thereby heating the atmosphere. The absorption...
or scattering of the solar radiation by atmospheric constituents is referred to as direct radiative effect, or aerosol-radiation interaction (ARI). The ARI due to BC has been shown to enhance global warming, especially in the Arctic region (AMAP, 2015; Collins et al., 2013; Sand et al., 2013, 2016).

In addition to ARI, deposited BC darkens both snow and ice cover and thereby reduces the albedo of reflective surfaces (AMAP, 2011). This in turn increases the amount of solar radiation absorbed by snow and ice, and diminishes the back-reflected portion of the solar insolation, which results in warming (Hansen & Nazarenko, 2004). Moreover, BC can impact the radiative balance indirectly by altering cloud properties through aerosol-cloud interactions and resulting rapid adjustments (Boucher et al., 2013). Pure BC particles are hydrophobic, and therefore inefficient as cloud condensation nuclei (CCN). However, when emitted to the atmosphere, BC particles typically undergo aging processes and become either coated or internally mixed with hydrophilic compounds. The resulting BC-containing particles are efficient as CCN (Dalirian et al., 2018), and, therefore, increasing aerosol number concentration through increased BC emission leads to increased cloud droplet number concentration. The cloud water is then distributed to a larger amount of particles, leading to smaller cloud droplets. This results in brighter clouds, which are more efficient in scattering SW radiation (Twomey effect, [Twomey, 1977]). The aerosol impacts on clouds are termed as indirect effects, or as aerosol-cloud interactions (ACI).

As BC is a very effective absorber of radiation, BC-containing particles can heat the atmosphere locally, which can affect cloud dynamics and atmospheric stability in the troposphere (Johnson et al., 2019). This local heating can also alter precipitation and convection at the regional scale, due to changes in atmospheric dynamics (Johnson et al., 2019). Therefore, BC-containing particles can impact the lifetime of clouds, cause cloud burn-off, and even change the planetary boundary layer (PBL) height by altering the vertical heating rate in the atmosphere (Ding et al., 2016). Apart from affecting cloud properties, a change in PBL height also can increase air pollution at the surface (Ding et al., 2016). As BC radiative forcing depends strongly on the altitude of BC particles (Ban-Weiss et al., 2012; Flanner, 2013), the surface temperature responses to changes in atmospheric BC concentrations are highly complex (Yang et al., 2019).

Along BC, combustion processes release various other aerosol compounds to the atmosphere. Here we consider OC and sulfate since they are the most common co-emitted aerosol species of BC (Lamarque et al., 2010), and are known to have strong radiative effects. In contrast to BC, both OC and sulfate particles are known to cool the atmosphere, due to the scattering of solar radiation back to space (ARI). Like BC, sulfate and OC have the ability to alter clouds, increasing their cooling potential (Boucher et al., 2013). In many regions, these negative radiative forcings of sulfate and OC have been found to out-weigh the positive radiative forcing of BC due to ARI of BC (Chen et al., 2010; Kühn et al., 2020). Although the combined effects of aerosol particles are generally thought to result in a cooling of the atmosphere, they are still quite poorly understood and many of the estimates include substantial uncertainty (Koch & Del Genio, 2010; Stjern et al., 2017; Yang et al., 2019). For instance, Bellouin et al. (2020) estimated a total effective radiative forcing (ERF) due to anthropogenic aerosols of $-1.6$ to $-0.6$ W m$^{-2}$, with a 90% confidence interval, whereas the ERF for well-mixed greenhouse gases has been estimated to lie between $2.3$ and $3.4$ W m$^{-2}$ (Myhre, Shindell, et al., 2013).

Whether aerosol particles will have an overall warming or cooling effect locally depends on various other factors, for example, on the spatial and vertical location of the aerosol, since the magnitude and sign of the aerosol radiative effect depend on surface and cloud characteristics. Furthermore, differences in the composition of emitted aerosol and oxidative conditions can have a strong influence on the radiative effects of aerosol particles (Paulot et al., 2018). Therefore, the climatic impacts of aerosol emissions can differ greatly between different countries (Aamaas et al., 2016). In contrast, the warming effect of well-mixed greenhouse gases is known to be somewhat independent of the source location. This means that while mitigating for example, CO$_2$ emissions from different countries will impact the global mean radiative balance at similar efficiency, the same does not apply for aerosols.

The aim of this study was to examine the radiative effects due to the removal of the anthropogenic aerosol emissions from Chile and Mexico. As a part of the multidisciplinary ERC project ClimaSlow (2017), this study was motivated by the national climate programs of the two countries. Both Chile and Mexico have announced ambitious goals for BC mitigation: Mexico declared a mitigation target of 51% BC emission
reduction by the year 2030 (SEMARNAT & INECC, 2016). Similarly, being one of the first nations, Chile has included SLCF mitigation in its national determined contribution (NDC) for the Paris agreement and has recently announced the unconditional aim of reducing its BC emissions by 25% by 2030 (Chile's Nationally Determined Contribution (NDC), Update 2020, 2020). Moreover, Chile and Mexico are interesting targets of analysis due to their closeness to the equator, as insolation is very strong in this region, which magnifies the radiative effects of aerosols. Furthermore, the climate of the Northern coast of Chile is partly dominated by a semi-persistent stratocumulus (Sc) deck (Abel et al., 2010; Huneeus et al., 2006; Klein & Hartmann, 1993; Wood et al., 2011). Stratocumulus clouds are low, shallow clouds that often occur in large, persistent sheets (decks) over relatively pristine, maritime environments (Stevens et al., 2013). Due to their structure, these clouds usually have lower albedo than thicker water clouds. Low albedo and the low number of CCN makes these stratocumulus clouds especially susceptible to aerosol-cloud interactions (Chen et al., 2015), as is for instance evident by the so-called ship tracks that are easily detectable in satellite imagery (Ackerman et al., 2000; Schreier et al., 2007). Due to this susceptibility, brightening of stratocumulus clouds through aerosol injection has even been suggested as a geoengineering method to slow global warming (Latham, 1990; Maalick et al., 2014). The Sc deck west of Chile has a significant impact on the Earth's radiation budget (Hartmann et al., 1992; Wood, 2012). A similar, but less persistent cloud deck exists over the Pacific, west of Mexico (Muhlbauer et al., 2014; Wood, 2012).

In this article, we studied the climatic effects due to removal of anthropogenic BC, OC and sulfur dioxide (SO₂) emissions that originate from Chile and Mexico, by using the global aerosol-climate model ECHAM-HAMMOZ (Kokkola et al., 2018; Schultz et al., 2018; Tegen et al., 2019) and the year 2015 anthropogenic aerosol emissions. While in the atmosphere, gaseous SO₂ typically becomes oxidized, eventually partitioning as sulfate into the particle phase. Therefore, reducing gaseous SO₂ emissions also decreases the amount of sulfate in aerosol particles. Our main focus was to analyze the radiative effects of mitigating these emissions and to determine how extensively aerosol-radiation interactions contribute to the total radiative effects. At first, we analyzed the areas where the anthropogenic emissions from Chile and Mexico contribute the most to the column integrated aerosol concentrations. This helped us identify regions where Chilean and Mexican aerosol has the largest potential to interact with radiation or to affect cloud properties. As the anthropogenic emissions analyzed here are quite small compared to the global total, we mainly restricted our analysis to the regions that are most affected by these emissions. To put the radiative effects of mitigating Chilean and Mexican emissions into global context and to demonstrate the differences in radiative response due to aerosol mitigation in different countries, we also compared them to the radiative effects due to reducing Chinese SO₂ emissions, which are one of the highest in the world for a single country (Li et al., 2017) and hence are very likely to produce a robust radiative forcing signal.

2. Methods

2.1. ECHAM-HAMMOZ

We conducted all simulations with the aerosol-climate model ECHAM-HAM2.3 (ECHAM-HAMMOZ). In ECHAM-HAMMOZ, the atmospheric general circulation model ECHAM (Roeckner et al., 2003; Stevens et al., 2013) is coupled with the aerosol module HAM which also takes into account the mixing of different compounds and atmospheric aging processes (Kokkola et al., 2018; Stier et al., 2005; Tegen et al., 2019). The HAM module includes the most relevant aerosol species: BC, OC, sulfate, mineral dust (DU) and sea salt (SS). Further, the detailed representation for aerosol is achieved by coupling the sectional SALSA2.0 aerosol micro-physics module (Kokkola et al., 2018) to the model.

SALSA discretizes the aerosol size distribution into 10 size sections, which provides an accurate and computationally efficient platform for resolving aerosol-atmosphere interactions (Kokkola et al., 2018). Furthermore, the micro-physical processes for each aerosol size class and species are solved separately for soluble and insoluble aerosol, simultaneously. A full description of the SALSA2.0 parametrization is presented in Kokkola et al. (2018). The grid resolution used for this study was T63L47, which corresponds to ~1.9° × 1.9° horizontal resolution, and 47 vertical layers up to 0.01 hPa (approximately 80 km altitude). The model version used in this study does not simulate albedo changes due to deposition of BC on snow and ice.
Cloud droplet activation is calculated according to the parametrization presented in Abdul-Razzak and Ghan (2002) which calculates the number of activated droplets for a given updraft velocity, aerosol size distribution, and composition. In SALSA2.0, BC is treated as an insoluble compound that can contribute to cloud activation in the Abdul-Razzak and Ghan scheme after becoming coated by condensing sulfuric acid (H$_2$SO$_4$) or becoming internally mixed with OC. Sulfur and OC are described as completely soluble compounds and the hygroscopicity values used in the micro-physics routines were 0.57 for sulfate and 0.21 for OC. A detailed description of treatment of different cloud processes and different radiative forcing mechanisms in ECHAM-HAMMOZ is presented in Neubauer et al. (2019).

In ECHAM-HAMMOZ, 97.5% of the anthropogenic sulfur emissions enter the atmosphere as gas (SO$_2$), while 2.5% of the SO$_2$ total emission mass is converted directly to sulfate particles (SO$_4^{2-}$). When SO$_2$ is oxidized in the atmosphere, it forms H$_2$SO$_4$, which then forms sulfate aerosol through new particle formation or condensation onto existing aerosol particles.

The representation of aerosol concentrations in the vertical scale in ECHAM-HAMMOZ coupled with SALSA2.0 has been validated against measurements as presented in Kokkola et al. (2018). Comparing aerosol optical depth (AOD) values to Moderate Resolution Imaging Spectroradiometer (MODIS) and AErosol RObotic NETwork (AERONET) data, they showed that AOD from simulations using ECHAM-HAMMOZ coupled with SALSA2.0 agrees reasonably well with the measured data over the regions west of Chile and Mexico. Furthermore, Kokkola et al. (2018) compared the vertical aerosol concentration profiles to the data from aircraft campaigns: the VAMOS Ocean-Cloud-Atmosphere-Land Study Regional Experiment (VOCALS-REx [Mechoso et al., 2014; Wood et al., 2011]) and CRE-AVE campaign (CRE-AVE, 2006), which were carried out west of Chile and south of Costa Rica. This comparison showed that the vertical concentration profiles for sulfate, BC and OC simulated with ECHAM-HAMMOZ coupled with SALSA2.0 are well in line with the measured values.

The validation of the cloud properties and aerosol-cloud interactions in ECHAM-HAMMOZ is presented in detail in Neubauer et al. (2019). Their analysis shows that in regions where shallow convective clouds are common, the ECHAM-HAMMOZ simulated cloud water and cloud cover are well in line with the observations. However, for these clouds the ECHAM-HAMMOZ derived cloud-top cloud droplet number concentration (CDNC) levels are too high, which might lead to overestimated SW cloud radiative effects in these regions. Furthermore, as it is typical for general circulation models, in ECHAM-HAMMOZ the SW cloud radiative effect is under-predicted in marine stratocumulus regions west of the continents (Neubauer et al., 2019). This in turn can result in over-prediction of SW radiation reaching aerosol below the cloud layer, affecting the modeled radiative response of aerosol emission mitigation. At the same time, ACI and the related radiative effects might be underestimated.

### 2.2. Anthropogenic Aerosol Emissions

For the global anthropogenic aerosol emissions of BC, OC and SO$_2$, we used the emissions for the year 2015 from the ECLIPSE V6a emission inventory (Z. Klimont, personal communication, February 14, 2020, Klimont et al., 2017), which was designed with the integrated assessment model GAINS (Amann et al., 2011). The BC and OC emissions from waste management sector were updated with the methodology described in Gómez-Sanabria et al., (2018). The spatial emission fields were re-gridded to the ECHAM-HAMMOZ model resolution (T63, approx. 1.9° × 1.9°). The anthropogenic emission strengths are presented in Table 1. As can be seen, the contributions from Chile and Mexico to the global total emissions are rather small: less than 0.6% for Chile, and 1.4% for Mexico.

The major sources of BC emissions are the domestic sector and traffic (Cruz-Núñez, 2014), and a large part of Mexican BC originates from urban metropolitan areas that are located on elevated territory. For instance, the altitude of Mexico City is over 2,200 m, and Guadalajara and Monterrey are located at altitudes of 1,500 and 500 m, respectively. In contrast, SO$_2$ emissions are distributed spatially more evenly within the industrial and the energy sectors. For Chile, Santiago and the regions south of Santiago are the most crucial BC and OC emission sources (Molina et al., 2015), whereas most SO$_2$ originates from northern Chile (Mena-Carrasco et al., 2014), with the strongest sources being the copper industry and energy production.
2.3. Experiment Design

In order to estimate the maximum radiative effect of mitigating anthropogenic aerosol emissions from Chile and Mexico, we conducted altogether seven simulations. In order to be able to study the effects of each substance individually, we performed separate simulations where always one of the substances was removed from the emissions inventory of Chile and Mexico. Additionally, to study the impacts of co-emitted species due to BC mitigation, we performed one further simulation where BC and OC emissions were reduced simultaneously. The differences between simulations carried out in this study are presented in Table 2.

Note that, because Chile and Mexico are separated by the inter-tropical convergence zone (ITCZ), most of the aerosol plumes originating from these two countries do not cross over to the respective opposite hemisphere and that their effects on the opposite hemisphere are negligible. We therefore assumed that the effects of Chilean and Mexican aerosol emissions can be studied individually using results from the same simulation (see Text S4 and Table S1).
The simulation FREE_WIND was used to generate wind fields to which all other simulations could be nudged. Nudging was used to reduce the uncertainties in radiative forcings due to modeled climate variability and differing meteorological fields, as will be discussed in Section 2.4. In FREE_WIND, the model meteorology was allowed to evolve freely and only sea surface temperature (SST) and sea ice cover (SIC) were fixed to climatological monthly mean values of the years 2000–2015, by using the results from the PCMDI’s (Program for Climate Model Diagnosis & Intercomparison) Atmospheric Model Inter-comparison Project (Taylor et al., 2012).

The global wind patterns and surface pressure fields for the rest of the simulations were nudged toward prescribed fields obtained from FREE_WIND using a Newtonian relaxation scheme (Zhang et al., 2014). Atmospheric temperature and dry static energy were allowed to evolve freely.

The reference simulation (BASE) included all aerosol emissions. Furthermore, we conducted three simulations that were otherwise identical to BASE but the anthropogenic aerosol emissions originating from Chile and Mexico were removed, separately for BC (NO_BC), OC (NO_OC) and SO₂ (NO_SO₂). Here we considered the emissions from agricultural waste burning to be a part of the anthropogenic emissions, but wildfire emissions were kept identical between the perturbed simulations and the BASE simulation.

Additionally, as BC and OC are usually co-emitted species, we also wanted to analyze the effects of reducing BC and OC simultaneously. To this end we performed a perturbed simulation (MITIG) where the anthropogenic BC and OC emissions from Chile and Mexico were simultaneously decreased by 40%. This was motivated by the Mexican and Chilean mitigation targets for BC. Additionally, our aim was to set the radiative effects of eliminating the Chilean and Mexican emissions into global context by comparing them to the effects of reducing Chinese SO₂ emissions, which are one of the highest in the world for a single country (Li et al., 2017). Here we used only SO₂ because the ratios of emissions of SO₂, OC, and BC were fairly similar for China, Chile, and Mexico and out of the three compounds, SO₂ showed the most robust radiative forcing signal. Therefore, we performed a similar simulation without the anthropogenic SO₂ emissions originating from China (NO_SO₂_China).

Each simulation was run for 11 years with the first year used as spin-up and the aerosol emissions following a monthly varying setup based on the emissions for year 2015. The forest fire and biomass burning emissions were taken from the GFAS emission inventory (Kaiser et al., 2012), using monthly varying climatologies, calculated based on the monthly mean values between the years 2000 and 2016. In all simulations, the SS and DU emissions were calculated online and were dependent on the 10 m wind speed. The SS emissions followed the parametrization of Guelle et al. (2001), while the DU emissions followed the parametrization of Tegen et al. (2002) which has been improved further by modifications by Cheng et al. (2008) and Heinold et al. (2016).

The emissions for the aviation sector were kept fixed for all simulations, and they were retrieved from the Emissions for Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) database (Lamarque et al., 2010) for the year 2015, using the Representative Concentration Pathway (RCP) 4.5 scenario (Thomson et al., 2011; van Vuuren et al., 2011). In addition, we used fixed ECLIPSE V6a CLE emissions for the year 2015 for the international shipping emissions in all of the simulations.

The greenhouse gas mixing ratios were set to values of the year 2010 in all of the simulations, following RCP 4.5. The mixing ratios for CO₂, CH₄ and nitrous oxide (N₂O) were set to values of 389.1 ppm, 1,767 ppb, 323 ppb, respectively. Furthermore, the ozone and OH mixing ratios were taken from reanalysis data (described in Inness et al. [2013]). The OH mixing ratios included diurnal variation which followed a cosine function between sunrise and sunset peaking at midday. For ozone, we used the monthly mean values for the year 2015, corresponding to the RCP scenario 4.5. Further details on how trace gas concentrations are treated in ECHAM-HAMMOZ are given in Stier et al. (2005) and Pietikäinen et al. (2014).

2.4. Radiative Forcing Calculations

In ECHAM-HAMMOZ, the radiative effect due to aerosol-radiation interactions (REₐₐ-router) is calculated according to Collins et al. (2006) by using a double-call with and without aerosol to the radiation calculation routine. We calculated the radiative forcing due to aerosol-radiation interactions (RFₐₐ-router) as the difference
in $\text{RF}_{\text{ARI}}$ between the perturbed simulation and the reference simulation (BASE) (Ghan, 2013; Neubauer et al., 2019). Note that this way of calculating $\text{RF}_{\text{ARI}}$ does not entirely follow the definition in Myhre, Samset, et al. (2013) for all-sky conditions, because the cloud properties between two simulations can change. However, the effect of these changes in cloud properties on the $\text{RF}_{\text{ARI}}$ are estimated to be small (Neubauer et al., 2019).

The total effect on the Earth’s radiation balance induced by a substance is estimated using the total effective radiative forcing ($\text{ERF}_{\text{ARI+ACI}}$) (Boucher et al., 2013; Lohmann et al., 2010; Mülmenstädt et al., 2019; Smith et al., 2018) which includes radiative forcing due to aerosol-radiation interaction ($\text{RF}_{\text{ARI}}$) and aerosol-cloud interaction ($\text{RF}_{\text{ACI}}$) as well as rapid adjustments to both radiative forcings (Boucher et al., 2013). We here calculated the $\text{ERF}_{\text{ARI+ACI}}$ for eliminating anthropogenic aerosol as a difference in net radiative flux at the top of the atmosphere (TOA) between a perturbed simulation and the reference simulation (BASE). This is usually done using simulations with a freely evolving meteorology, but with fixed SST and SIC (Boucher et al., 2013; Lohmann et al., 2010). However, if the radiative forcing is small, the modeled climate variability in yearly average radiative budget introduces substantial uncertainty in the calculated $\text{ERF}_{\text{ARI+ACI}}$, which may be larger than the actual signal itself (e.g., [AMAP, 2015; Forster et al., 2016; Kühn et al., 2020]). In order to reduce this uncertainty we here nudged the modeled wind fields and surface pressure in addition to keeping SST and SIC fixed (see Section 2.3). To make this additional constraint as clear as possible, we hereafter refer to this nudged $\text{ERF}_{\text{ARI+ACI}}$ as $\text{ERF}_{\text{NDG}}$.

Various studies have shown that nudging can affect some of the model processes which strongly depend on wind and precipitation levels, like, for instance, cloud formation and dust and sea salt emissions (Astitha et al., 2012; Lin et al., 2016; Sun et al., 2019). This may also affect the ERF values obtained in nudged simulations (Forster et al., 2016). In order to minimize the effect the nudging has on the modeled ERF values, we nudged here toward model-own (simulation FREE_WIND) derivatives of wind fields (vorticity and divergence) and surface pressure, and did not nudge the atmospheric model temperature at all. This has been shown to reduce the effects of nudging on the calculated ERF (Lin et al., 2016; Sun et al., 2019; Zhang et al., 2014). In summary, the $\text{ERF}_{\text{NDG}}$ values obtained in this study do not include all the possible rapid adjustments (Forster et al., 2016), and thus, do not correspond fully to the $\text{ERF}_{\text{ARI+ACI}}$ that is usually used (Boucher et al., 2013; Neubauer et al., 2019).

### 2.5. Determining the Area for Analysis

As the anthropogenic emissions from Chile and Mexico are small compared to the global total, the area that is significantly influenced by these emissions is also relatively small. Consequently, the radiative effects of these emissions are small on a global scale as well. However, their regional impact can still be important. We therefore identified the regions most affected by Chilean and Mexican aerosol emissions, both directly and indirectly.

To this end, we identified our regions of interest (ROI) by investigating changes in the areas with noticeable changes in vertically integrated aerosol and cloud droplet concentrations (burdens). A detailed description of the calculation of ROIs is presented in Text S1 and the changes in aerosol and cloud droplet number concentration (CDNC) burdens are presented in Figure S1. The resulting ROIs for Chile and Mexico are shown in Figure 1. Unless stated otherwise (e.g., Section 3.3.1), we restrict the following analysis to these two ROIs, which we will refer to as MexicoROI and ChileROI.

### 3. Results

In this section, we examine and compare the obtained changes in atmospheric aerosol concentrations and radiative fluxes separately for specific regions for Chile and Mexico. Finally, we compare the effects of reducing Chilean and Mexican $\text{SO}_2$ emissions on global $\text{ERF}_{\text{NDG}}$ to the effects of eliminating Chinese an-
thropogenic SO\textsubscript{2} emissions. All the results presented throughout this section represent the mean difference values of the area weighted mean values for the ROIs presented in Section 2.5. The averaging was done by first calculating the yearly averages for all the perturbed simulations, and then by computing the mean of yearly differences with respect to the reference simulation (BASE). The uncertainty intervals are presented as one standard deviation which was calculated also based on the differences of yearly mean values between perturbed and reference simulations. Furthermore, as most of the SO\textsubscript{2} is converted to sulfate, we study the effects of reducing SO\textsubscript{2} on clouds and radiation by analyzing the changes in sulfate concentrations.

In addition, as the SST values were fixed in our simulations, they mostly dictate the surface temperature and the effects of aerosol on surface temperatures were very small. The effects of aerosol on precipitation were also small and they were accompanied with large modeled variability. Therefore, we did not analyze the effects of aerosol reductions on the precipitation levels.

### 3.1. Atmospheric Aerosol Concentrations

By examining the differences in aerosol mass burdens for each perturbed simulation, we can estimate how much both countries contribute to the aerosol load over their respective ROIs. In addition to the total mass burden, the horizontal and vertical distribution of the particles, especially with respect to the cloud layer, determines their impacts on the radiative balance. The average differences between the perturbed simulations and the reference simulation (BASE) in aerosol mass burden for BC, OC and sulfate (averaged over the entire simulation period and the respective ROI area) are shown in Table 3.

| Burden (µg m\textsuperscript{-2}) | Chile\textsubscript{ROI} | Mexico\textsubscript{ROI} | Global |
|-----------------------------------|--------------------------|---------------------------|--------|
| NO\textsubscript{BC}: BC          | −24.1 ± 1.9 ((−4.9 ± 0.4)%) | −76.0 ± 3.9 ((−12.3 ± 0.6)%) | −6.5 ± 0.6 ((−0.8 ± 0.1)%) |
| NO\textsubscript{OC}: OC          | −66.4 ± 23.8 ((−2.0 ± 0.7)%) | −172.8 ± 13.8 ((−4.8 ± 0.4)%) | −16.8 ± 7.2 ((−0.3 ± 0.1)%) |
| NO\textsubscript{SO\textsubscript{2}}: sulfate | −158.0 ± 14.1 ((−3.8 ± 0.3)%) | −271.0 ± 20.1 ((−5.7 ± 0.4)%) | −28.1 ± 4.4 ((−0.6 ± 0.1)%) |
| MITIG: BC                         | −10.5 ± 1.5 ((−2.1 ± 0.3)%) | −30.7 ± 1.7 ((−4.9 ± 0.3)%) | −2.8 ± 0.4 ((−0.3 ± 0.1)%) |
| MITIG: OC                         | −32.6 ± 15.9 ((−1.0 ± 0.5)%) | −72.6 ± 8.5 ((−2.0 ± 0.2)%) | −9.9 ± 3.8 ((−0.2 ± 0.1)%) |

**Note.** The uncertainty values are calculated as one standard deviation of the yearly difference values. Relative differences are shown in parentheses.

Even though we restricted our analysis to Chile\textsubscript{ROI} and Mexico\textsubscript{ROI}, the changes in BC, OC and sulfate burdens due to the removal of the anthropogenic emissions of these substances in Chile and Mexico are fairly small. This is mostly due to transportation of aerosol and emissions from other countries. Furthermore, in order to obtain ROIs that are as large as possible, we chose very small threshold values in order to maximize the ROI area, which directly affects the average burden changes in the ROIs. Due to differences in orography, meteorology and horizontal distribution of the emissions in Chile and Mexico, the aerosol particles in Chile\textsubscript{ROI} and Mexico\textsubscript{ROI} are transported differently with in the respective ROIs as well.

The Chilean anthropogenic emissions seem to make a fairly small contribution to the BC and sulfate burden values over Chile\textsubscript{ROI}, and even smaller to the OC burden. For Chile\textsubscript{ROI}, we found that the total BC burden changes by (−4.9 ± 0.4)%; whereas for OC the change is only (−2.0 ± 0.7)%. The sulfate burden also changes by (−3.8 ± 0.3)%. For scenario MITIG, the decrease in the BC and OC burdens is in line with the emission reductions: the change in BC burden ([−2.1 ± 0.3]%) is almost half of the change observed for NO\textsubscript{BC} simulation ([−4.9 ± 0.4]%), and similarly for OC burden ([−1.0 ± 0.5]% versus [−2.0 ± 0.7]%).

As for Chile, the emissions from Mexico make a rather small contribution on the sulfate and OC burden over Mexico\textsubscript{ROI}. The burden changes over Mexico\textsubscript{ROI} are, however, larger than over Chile\textsubscript{ROI} in both absolute and relative terms (see Table 3). With (−76.0 ± 3.9) µg m\textsuperscript{-2}, which corresponds to (−12.5 ± 0.6)%, the change in BC burden over Mexico\textsubscript{ROI} is substantial.
All in all, it appears that the relative changes in average burden values are slightly larger for MexicoROI than for ChileROI. One explanation for this is the differences in spatial distribution of anthropogenic aerosol emissions: Huneeus et al. (2006) state that the main Chilean SO$_2$ emitters (i.e., copper smelters) are located in the northern part of Chile, from where the emitted sulfate is transported north and northwest and mainly remains at altitudes below 4 km. The sources for BC and OC are mostly in the middle, near the capital region. This causes that the changes in the number of particles with diameters larger than 100 nm ($N_{100}$) are also more spread toward both north and south, resulting in relatively larger ROI for Chile, as shown in Figure S1. In Mexico, the emission sources are also distributed differently along the country for sulfur and BC, but they are not as distinct as for Chile, and all of the emissions are distributed more evenly across the ROI.

3.2. Aerosols and Clouds

As indicated in Section 1, one of the motivations for this study was to analyze the effects of anthropogenic aerosol emissions on the maritime stratocumulus decks off the Chilean and the Mexican coasts. However, the current configuration for ECHAM-HAMMOZ does not provide output diagnostics for determining different cloud types directly. Hence, we examined the vertical profiles of the annual mean water cloud fraction, using the data from the reference simulation (BASE). Since typical maritime Sc clouds have cloud bases below 2 km and a cloud thickness of less than 1 km (Wood, 2012), we identified grid boxes where the maximum annual mean water cloud fraction was below 900 hPa, and determined those as the areas which are dominated by maritime Sc clouds in our model. These grid boxes are marked in Figure 2, which also shows how much the defined ROIs coincide with these Sc cloud decks.

Besides horizontal distribution, the vertical distribution of aerosol particles and clouds affects the radiative properties. For instance, aerosol particles above cloud layer receive a higher portion of SW radiation than below clouds since clouds are efficient in reflecting SW radiation. This is important for absorbing aerosol, like BC, whose warming effect is often enhanced on top of a cloud (Kühn et al., 2014; Zarzycki & Bond, 2010). Furthermore, in order for aerosol-cloud interactions to occur, aerosol particles and cloud droplets need to coincide both vertically and horizontally. Because of the differences in local conditions (e.g., atmospheric circulation) for ChileROI and MexicoROI, the vertical distributions for aerosol compounds and clouds are studied separately for the two ROIs.
For the Chilean anthropogenic emissions, the total mass of SO₂ emitted yearly is remarkably larger than the mass of BC or OC. This leads to a high contribution to atmospheric aerosol number concentrations, which further affects the CDNC. Besides the total mass, the hygroscopicity of sulfate is higher than for OC and BC, and the particle size ranges in which these substances occur, differ (not shown). Altogether these differences lead to changes in SO₂ emissions having a much stronger effect on CDNC than BC and OC do, as we discuss later on in Section 3.2.2. The number of particles with diameters larger than 100 nm (N₁₀₀) is typically considered a proxy for particles that can potentially act as CCN (Dusek et al., 2006; Janssen et al., 2011; Tröstl et al., 2016). The vertical concentration difference profiles between perturbed and reference simulation (BASE) for BC, OC and sulfate mass, N₁₀₀, and in-cloud CDNC for ChileROI are presented in Figure 3. In order to illustrate the changes in aerosol concentrations with respect to the water cloud layer, we also show the annual average in-cloud CDNC and liquid water content (LWC) from the reference simulation (BASE) in the two rightmost panels. Here the in-cloud CDNC represents the CDNC values that are weighted with liquid cloud time fraction, whereas the LWC is the non-weighted, average value.

The vertical location of the atmospheric BC aerosol particles, especially with respect to the clouds, strongly affects BC radiative forcings. Figure 3a shows that most of the changes in BC concentration are at or below cloud level, that is, below the pressure levels where the LWC and in-cloud CDNC are at their maximum for the BASE simulation (Figures 3f and 3g). The same applies to the changes in OC and sulfate concentrations (Figures 3b and 3c). As most of ChileROI is covered with a persistent Sc deck, it can thus be expected that the magnitude of the radiative effects is reduced compared to a situation where BC resides above the cloud.

Figure 3d shows the changes in N₁₀₀ for the different scenarios. The sulfur emissions clearly have the strongest effect on N₁₀₀. Furthermore, due to its high hygroscopicity (Petters & Kreidenweis, 2007), sulfur-induced N₁₀₀ changes affect CDNC more than similar changes due to BC and OC. Furthermore, the vertical concentration difference profile of N₁₀₀ for NO_SO₂ decreases almost linearly with altitude, while in the other scenarios the decrease is much more rapid with height. It can therefore be expected that the magnitude of the cloud effects due to sulfate aerosol is strong compared to the effects due to BC and OC.

Note that the graphs in Figure 3 represent the mean concentrations for model hybrid levels, and the hybrid pressure level values are the ROI-averaged values for each model level. In order to analyze how the surface emissions from elevated altitudes affect concentrations, we calculated the mean concentration also as a function of actual pressure for 25 pressure intervals by binning grid boxes of similar pressure and averaging over the values. These results for ChileROI are presented in Figure S3. Figure S3 shows a local minimum for the BC and OC concentration changes below 800 hPa, which coincides well with the local maximum

3.2.1. ChileROI

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Note that the graphs in Figure 3 represent the mean concentrations for model hybrid levels, and the hybrid pressure level values are the ROI-averaged values for each model level. In order to analyze how the surface emissions from elevated altitudes affect concentrations, we calculated the mean concentration also as a function of actual pressure for 25 pressure intervals by binning grid boxes of similar pressure and averaging over the values. These results for ChileROI are presented in Figure S3. Figure S3 shows a local minimum for the BC and OC concentration changes below 800 hPa, which coincides well with the local maximum
in the CDNC profile of the BASE simulation in the same figure. This further supports our interpretation that a large part of the Chilean BC is in or below cloud. Furthermore, we present the spatial distributions of changes in Chilean anthropogenic aerosol concentrations at 970, 900, 800, and 750 hPa pressure levels in Figure S5. This figure visualizes quite well how the anthropogenic aerosol plume originating from Chile is transported first north and then westward along the South-American coast while it is slowly lifted upwards.

As removing anthropogenic aerosol decreases the atmospheric aerosol concentrations and thereby the CCN concentrations, the CDNC decreases as well (Figures 3d and 3e). Correspondingly, the mean effective radius of cloud droplets increases when the aerosol burden is decreased (not shown). However, the maximum difference in the effective radius can be observed at 970 hPa with less than 0.1 µm ([0.7 ± 0.3]%) increase for the NO_SO2 simulation, which is a relatively small change. We will show later in Section 3.3 that the aerosol-cloud interactions affect radiative balance remarkably stronger than the radiative forcing due to aerosol-radiation interactions for ChileROI.

Since the aerosol emission reductions lead to larger cloud droplets, the remaining droplets precipitate more easily due to increased collision coalescence efficiency. This can be observed as decreasing mean cloud water content (not shown). However, this decrease is relatively small and is accompanied by a large uncertainty. The maximum average change in vertically integrated cloud water over ChileROI is (−0.86 ± 0.25) g m⁻² (−1.68 ± 0.48[%]) between NO_OC and BASE simulations, whereas for NO_BC the change is (−0.30 ± 0.16) g m⁻² (−0.59 ± 0.30[%]) and for NO_SO2, the difference to reference simulation (BASE) is (−0.73 ± 0.16) g m⁻² (−1.42 ± 0.31[%]). Although this cloud lifetime effect (Albrecht, 1989) is included in the model physics, it is still highly uncertain and is speculated to have a minor role compared to the cloud albedo effect (Malavelle et al., 2017). As the simulated changes in cloud water are very small, indicating that the cloud lifetime effect will also have a minor role in the obtained total radiative effect of aerosol reductions, we did not analyze this effect further.

3.2.2. MexicoROI

The vertical concentration difference profiles between perturbed and reference simulation (BASE) for BC, OC, and sulfate mass, N₁₀₀ and in-cloud CDNC for MexicoROI are presented in Figure 4. Like for ChileROI, the annual mean in-cloud CDNC concentration and the LWC for the BASE simulation are included in the panels at the right. As can be seen from Figures 3 and 4, the height of the cloud layer and vertical aerosol concentration profiles are different for MexicoROI and ChileROI.

In contrast to ChileROI, the anthropogenic BC concentration difference has a second minimum at the altitude of approximately 750 hPa, which is well above the maximum in-cloud CDNC of the liquid cloud layer (Figure 4f). The LWC for the BASE simulation has two maxima, one at 910 hPa, coinciding with the maxi-
maximum in in-cloud CDNC, and one at 720 hPa (Figure 4g), indicating that water clouds in MexicoROI extend to much higher altitudes compared to ChileROI. However, at 720 hPa the corresponding in-cloud CDNC value for the BASE simulation is much smaller than at 910 hPa. This indicates that the clouds around 720 hPa are less reflecting than the clouds below, and therefore they allow SW radiation to reach lower altitudes where the BC particles reside. This is visible even better in Figure S4, which has been prepared in the same manner as Figure S3. Here the vertical profile in anthropogenic BC concentration difference shows a clear minimum at 800 hPa, while the CDNC values of the BASE simulation peak at 900 hPa. As BC light absorption is enhanced above clouds, this can be expected to lead to a difference in radiative forcings between ChileROI and MexicoROI. The vertical profiles for OC and sulfate (Figures 4b–4c) have a second minimum at about 750 hPa as well, which manifests at 800 hPa in Figure S4.

While in ChileROI the SO2 emissions affect N100 most, the OC emissions from Mexico affect the N100 levels more than SO2. However, sulfur emissions have a stronger effect on CDNC than BC and OC emissions, even though the effect is not as noticeable as for ChileROI. Since the hygroscopicity of OC is lower than that of sulfur (Petters & Kreidenweis, 2007), the OC particles do not form cloud droplets as effectively as sulfate. That is why the differences in N100 levels do not directly correspond to the differences observed for CDNC, and thus the Mexican sulfur emissions affect the CDNC values more than the OC emissions. The changes for cloud droplet radius are of the same order of magnitude as for ChileROI, including high uncertainty (not shown).

The vertical concentration profiles for ChileROI and MexicoROI differ because of the differences in the ROIs (e.g., total area, spatial distribution of emissions), and also due to the differences in atmospheric circulation patterns, solar insolation, precipitation levels (Müllménstäd et al., 2020; Sierra et al., 2015) and aerosol transport. Furthermore, a large part of the Mexican aerosol emissions are released at rather high altitudes, since one of the major source regions, Mexico City, is located at 2,200 m. This can be observed also from Figure S4 which shows a clear peak in anthropogenic BC concentration differences around 800 hPa. In Figure S6 we further present the spatial distributions of changes in Mexican anthropogenic aerosol concentrations at 970, 900, 800, and 750 hPa pressure levels. This figure visualizes quite well the uplift and westward transport of the anthropogenic aerosol plume originating from Mexico.

### 3.3. Radiative Forcing

The differences in the aerosol mass burdens and the vertical concentration profiles between ChileROI and MexicoROI ultimately also lead to different radiative forcings for the two ROIs. Figure 5 shows the radiative forcing due to aerosol-radiation interactions (RFARI) and the ERFDG for the perturbed simulations. Note
that these radiative forcings are due to the removal of aerosol emissions, which leads to positive radiative forcings for cooling agents and negative radiative forcings for warming agents. In order to get an estimate of the combined strength of the contribution of RFARI and the rapid adjustments to ERFNDG, we visualize also the difference between ERFNDG and RFARI. The numerical values of RFARI and ERFNDG for ChileROI and MexicoROI are presented in Table 4. For all simulations, the 2D ERFNDG signal due to the emission perturbations is well covered by the ROIs defined in Section 2.5 (see Figure S7).

As shown earlier in Figures S1 and 2, the sulfate particles from Chile are mostly transported to the maritime stratocumulus region. This explains what we can observe from Figure 5: the RFARI for the NO_SO2 simulation is only slightly positive for ChileROI, while the ERFNDG is the largest in all scenarios and both ROIs. In this region, the RFARI for sulfate is reduced by clouds, but the cloud effects, especially the Twomey effect, are strong. As BC is a strong absorber, the removal of anthropogenic BC in the NO_BC simulation produces a negative RFARI. Yet, as discussed in Section 3.2, the anthropogenic BC aerosols are primarily found within or below cloud, leaving a fairly small all-sky BC RFARI for ChileROI when analyzing the effects of BC mitigation. Similarly, as for the Chilean sulfur emissions, the RFARI for the removal of OC is negligible (0.00 ± 0.01 W m⁻²). Although the area where we can observe the largest decreases of OC burden is outside of the stratocumulus deck at the Chilean coast (Figure 2), most of the OC aerosol is masked by higher-level clouds.

The resulting ERFNDG values due to removing aerosol emissions for ChileROI are positive for all the perturbed simulations, but include wider uncertainty ranges than the corresponding RFARI values. As observed in Section 3.2, the changes in CDNC levels are highest for NO_SO2, which translates to a large ERFNDG, with local maximum values of about 3 W m⁻² (see Figure S2). The ERFNDG signal for NO_OC is also positive (0.3 ± 0.1 W m⁻²), as the changes in CDNC close to the surface are the second largest of all scenarios (see Figure 3). The ERFNDG for the NO_BC simulation, on the other hand, is the smallest of all scenarios, which is due to the negative RFARI and much smaller changes in cloud properties.

For MexicoROI, the RFARI for the NO_SO2 simulation is only slightly positive, and like for ChileROI, for NO_OC the RFARI is negligible. In contrast to NO_OC and NO_SO2, the RFARI for NO_BC is considerably negative, that is, removing BC induces cooling of the atmosphere when analyzing only the aerosol-radiation interactions. In general, the BC radiative forcing due to ARI is known to be more efficient when BC particles are located above highly reflecting surfaces, such as clouds, which in contrast to dark, absorbing surfaces such as oceans. As discussed in Section 3.2, the portion of BC emitted from Mexico that ends up above cloud is considerably larger than that emitted from Chile (Figures 3 and 4), which can partly explain why the RFARI for NO_BC is larger for MexicoROI than for ChileROI. In order to analyze this further, we visualized the NO_BC RFARI separately for no-cloud (clear sky) and cloudy (all-sky) conditions (Figure S8). The difference between all-sky and clear sky RFARI is shown in Figure S8c, highlighting the effect of clouds on NO_BC RFARI. At the west of Mexico, in the region of the maritime Sc deck, the NO_BC all-sky RFARI is more negative. This means that a large part of the change in BC concentration happens above optically thickest part of the cloud, where the cooling effect of the BC removal is enhanced. Together with the fact that the changes in BC burden in MexicoROI are approximately three times larger than in ChileROI, this explains the higher RFARI in Mexico quite well.

Table 4
The Mean ROI-Averaged Radiative Forcing due to Aerosol-Radiation Interactions (RFARI) and ERFNDG at the TOA for ChileROI and MexicoROI

| Simulation | RFARI (W m⁻²) | ERFNDG (W m⁻²) |
|-----------|--------------|----------------|
|           | ChileROI     | MexicoROI      | ChileROI     | MexicoROI      |
| NO_SO2    | 0.00 ± 0.01  | 0.03 ± 0.01    | 0.48 ± 0.11  | 0.27 ± 0.06    |
| NO_BC     | −0.04 ± 0.01 | −0.11 ± 0.01   | 0.11 ± 0.14  | −0.10 ± 0.10   |
| NO_OC     | 0.00 ± 0.01  | 0.00 ± 0.01    | 0.30 ± 0.13  | 0.21 ± 0.10    |
| MITIG     | −0.02 ± 0.01 | −0.05 ± 0.01   | 0.23 ± 0.09  | 0.08 ± 0.11    |
| 0.4*NO_BC+0.4*NO_OC | −0.01 ± 0.01 | −0.05 ± 0.01   | 0.16 ± 0.11  | 0.04 ± 0.08    |
While in ChileROI removing anthropogenic BC emissions (NO_BC) causes positive ERF\textsubscript{NDG} values, the ERF\textsubscript{NDG} values for removing BC in MexicoROI are negative ($(-0.10 \pm 0.10)$ W m\textsuperscript{-2}). This is because here the RF\textsubscript{ARI} is much stronger and the radiative effects due to aerosol-cloud interactions and rapid adjustments are smaller than in ChileROI. However, the standard deviation is noteworthy. For NO\textsubscript{SO2}, the ERF\textsubscript{NDG} is positive with the value of $(0.27 \pm 0.06)$ W m\textsuperscript{-2}, and the variance is smaller than for NO\textsubscript{BC}. As for ChileROI, over MexicoROI the cloud effects and rapid adjustments are dominating for as a result of removing anthropogenic OC emissions (NO\textsubscript{OC}), with positive ERF\textsubscript{NDG} values of $(0.21 \pm 0.10)$ W m\textsuperscript{-2}.

The radiative effects for ChileROI and MexicoROI differ partly due to unequal ROIs: the ROI defined for Mexico covers relatively more ocean than ChileROI, which causes the mean surface albedo of MexicoROI to be smaller than for ChileROI. As pointed out in Section 3.2.2, the differences in, for instance, local meteorology and solar insolation levels will also affect the radiative responses for the emission reductions, resulting in differing ERF\textsubscript{NDG} values for ChileROI and MexicoROI. In addition, ChileROI is in general cloudier than MexicoROI. Clouds mask some of the incoming SW radiation, reducing the solar radiation reaching the aerosol particles. This results in smaller RF\textsubscript{ARI} values for ChileROI compared to MexicoROI. Inversely, aerosol-cloud interactions are stronger for ChileROI than for MexicoROI, evident through the higher ERF\textsubscript{NDG} values for ChileROI compared to MexicoROI. This can to the largest part be explained with the existence of the maritime Sc deck west and northwest of Chile, which is very susceptible to changes in aerosol concentrations (Bulatovic et al., 2019; Costantino & Bréon, 2013; Toll et al., 2017). Furthermore, ChileROI has a higher fraction of ocean surface, with sulfur emissions mostly in the northern part of the country, and BC and OC emissions in the middle. This causes the sulfate particles to be transported north where there is more incoming SW radiation, while BC and OC move more toward the east. In Mexico, the aerosol species are transported mainly to the same direction.

As in MITIG simulation both BC and OC emissions are reduced simultaneously by 40%, it is to be expected that the radiative forcing values somehow reflect the radiative forcing values of both the NO\textsubscript{BC} and NO\textsubscript{OC} simulations. The RF\textsubscript{ARI} values in MITIG are, to the accuracy of one standard deviation, linear combinations of the RF\textsubscript{ARI} values of NO\textsubscript{BC} and NO\textsubscript{OC} (see Table 4). However, the obtained ERF\textsubscript{NDG} values in MITIG cannot be approximated in the same fashion. As it can be seen from Table 4, the relation $0.4\text{ERF}_{\text{NDG,NO\textsubscript{BC}}} + 0.4\text{ERF}_{\text{NDG,NO\textsubscript{OC}}}$ is not equal with the ERF\textsubscript{NDG} values for the MITIG simulation. This demonstrates the complex interplay between aerosol-radiation and aerosol-cloud effects of different, co-emitted aerosol species, which has also been reported in other studies (Chen et al., 2010; Huang et al., 2018; Wang et al., 2015). However, since the ERF\textsubscript{NDG} for NO\textsubscript{OC} is positive and much greater than for the NO\textsubscript{BC} simulation, the overall ERF\textsubscript{NDG} for the MITIG simulation is also positive for both ChileROI and MexicoROI, despite the negative ERF\textsubscript{NDG} obtained for NO\textsubscript{BC} in MexicoROI. Like in Huang et al. (2018) and Kühn et al. (2020), our results underline the importance of including the reductions of co-emitted species when analyzing the effects of BC mitigation.

One remarkable feature is the dominant contribution of the aerosol-cloud interactions and rapid adjustments to the ERF\textsubscript{NDG} in ChileROI due to the removal of BC. On the other hand, the difference between ERF\textsubscript{NDG} and RF\textsubscript{ARI} for NO\textsubscript{BC} was small for MexicoROI. This is most likely due to the combination of various processes, for example, smaller Twomey effect, more BC above cloud and BC-induced changes in the heat balance of the cloud layer. Even though the simulations in this study where nudged in order to minimize differences between the model runs due to differences in the model meteorology, the ERF\textsubscript{NDG} standard deviations are still quite notable. This might be partly due to the rather small analyzed area, and the fact that the anthropogenic aerosol emissions of Chile and Mexico are quite small compared to the sum of all natural and anthropogenic sources affecting ChileROI and MexicoROI. Of all variables analyzed, the changes in cloud properties showed the largest standard deviations. Determining how much individual processes contribute to the ERF\textsubscript{NDG} was beyond the scope of this study.

### 3.3.1. Impact of the Location of Emissions Reductions

Contrary to the radiative effects of well-mixed greenhouse gases, such as CO\textsubscript{2}, aerosol radiative effects show much stronger horizontal and vertical variability, mostly due to the much shorter atmospheric lifetime of aerosols (Boucher et al., 2013) and differences in the regional cloud cover. In addition, aerosol particles emitted in the mid-latitudes are likely to induce a different radiative forcing compared to particles from...
regions near the equator (Kühn et al., 2014; Laakso et al., 2017), because average solar insolation is strongest at the equator. In order to set the ER\textsubscript{NDG} for removing the Chilean and Mexican SO\textsubscript{2} emissions into global context, we compare them to the ER\textsubscript{NDG} for reducing Chinese anthropogenic SO\textsubscript{2} emissions. Here we chose China because its size and high aerosol emissions are most likely to produce a robust radiative forcing signal, even though meteorological conditions are not fully comparable to Mexico and Chile. For this purpose, we defined the global radiative forcing efficiency, EFF\textsubscript{ERF}, as the ratio between the global ER\textsubscript{NDG} (i.e., averaged over the entire globe) and the total emission reduction in a country. The global ER\textsubscript{NDG} and calculated EFF\textsubscript{ERF} values for NO\textsubscript{SO2} and NO\textsubscript{SO2}\textunderscore{china} simulations are presented in Table 5.

Apart from China being located further away from the equator, the Chinese aerosol-cloud interactions are presumably more saturated (Boucher et al., 2013; Kühn et al., 2014) due to much higher aerosol burdens there. For warm clouds, the fraction of aerosol activated as CCN decreases with increasing aerosol number concentration, as relatively fewer particles become activated (Verheggen et al., 2007). This means that the emission reductions from China are expected to be less efficient than from countries where the aerosol emissions are smaller. This is also the case when analyzing the simulation data. SO\textsubscript{2} reductions from Chile and Mexico show a 20 times higher EFF\textsubscript{ERF} compared to reductions in China.

However, the global mean ER\textsubscript{NDG} for Chile and Mexico has a relatively high standard deviation compared to the central value (\(0.027 \pm 0.021\) W m\(^{-2}\)), whereas for emission reductions in China we observe a more detectable signal of (\(0.124 \pm 0.023\) W m\(^{-2}\)). This due to China's larger total aerosol emissions. Nevertheless, the obtained results underline the point that the location of aerosol emission mitigation matters when analyzing the global radiative balance.

### 4. Discussion and Conclusions

The purpose of this study was to demonstrate how emission mitigation in different countries and regions can have diverse, and even contrasting potential to affect the Earth’s energy budget. The motivation for this study stemmed from the current discussion on the potential of short-lived climate forcer (SLFC) mitigation to slow down global warming. We focused on analyzing the climatic effects of removing anthropogenic black carbon (BC), organic carbon (OC) and sulfur dioxide (SO\textsubscript{2}) originating from Chile and Mexico. These two Latin American countries have released ambitious climate programs that also consider SLFC emissions, including even specified reduction levels for their BC emissions. In addition, the Chilean local climate is strongly influenced by a persistent stratocumulus cloud deck west of Chile, which makes it an interesting target for studying aerosol-cloud interactions with large scale climate models. A similar, but less persistent cloud deck also exists west and northwest of Mexico.

For distinguishing the effects of different aerosol species, we compared perturbed cases without anthropogenic emissions of BC, OC and SO\textsubscript{2} originating from Chile and Mexico (NO\textsubscript{BC}, NO\textsubscript{OC} and NO\textsubscript{SO2}, respectively) against a reference simulation with all aerosol species present (BASE). Furthermore, in order to study the effect of co-emitted species of BC, we performed one further simulation (MITIG) with Mexican and Chilean anthropogenic BC and OC emissions reduced by 40%. The results were then analyzed for specific regions of interest (Chile\textsubscript{ROI} and Mexico\textsubscript{ROI}), defined for the areas where we could observe notable changes in the aerosol and cloud properties. As these ROIs were relatively small, we nudged the wind and surface pressure in all simulations to the same, model-generated wind and surface pressure fields.

For both Chile\textsubscript{ROI} and Mexico\textsubscript{ROI}, the obtained effects due to aerosol-cloud interactions and rapid adjustments were much stronger than the radiative forcings due to aerosol-radiation interactions. When removing BC, which strongly absorbs solar radiation, we obtained negative radiative forcing from aerosol radiation interaction, RF\textsubscript{ARI}. Eliminating anthropogenic sulfur emissions resulted in small positive RF\textsubscript{ARI}, since the sulfur particles are known to back-scatter incoming solar radiation. However, the magnitude of this effect

### Table 5

| Simulation       | Global ER\textsubscript{NDG} (W m\(^{-2}\)) | SO\textsubscript{2} reduction (kt yr\(^{-1}\)) | EFF\textsubscript{ERF} (W m\(^{-2}\)/(kt yr\(^{-1}\))) |
|------------------|------------------------------------------|---------------------------------------------|--------------------------------------------------|
| NO\textsubscript{SO2} | 0.027 ± 0.021                           | 873.67                                      | 3.086 × 10\(^{-5}\)                              |
| NO\textsubscript{SO2}\textunderscore{china} | 0.124 ± 0.023                           | 15018.67                                    | 0.154 × 10\(^{-5}\)                              |

Note. The efficiency EFF\textsubscript{ERF} is defined as a ratio of global ER\textsubscript{NDG} and total SO\textsubscript{2} emission reduction.
was much smaller than for removing the BC emissions. For the NO_OC simulation, the RFARI was virtually negligible.

In order to study the total effects on radiation due to removal of aerosol in perturbed simulations, we calculated the effective radiative forcing (ERF_{NDG}), which includes radiative forcing for both aerosol-radiation interactions and aerosol-cloud interactions, as well as rapid adjustments due to aerosol concentration perturbations. As our simulations where nudged, however, the simulations do not consider some adjustments. For NO_SO2 and NO_OC, ERF_{NDG} was positive for both ChileROI and MexicoROI, as expected, dominated by the aerosol-cloud interactions and rapid adjustments. The NO_BC case is more interesting, however, as the magnitudes of the RFARI and the forcing due to aerosol-cloud interactions and rapid adjustments are roughly of the same order but opposite in sign. This resulted in a positive ERF_{NDG} for ChileROI and a negative ERF_{NDG} for MexicoROI, both values also having an uncertainty that is roughly of similar magnitude as the estimate itself. This result highlights the difficulty in anticipating the global ERF caused by BC mitigation without simulating the effects of co-emitted species.

Due to different characteristics of the local climates and especially cloud layers, the ERF_{NDG} signals differed between ChileROI and MexicoROI. These features include, for instance, local cloud cover, land orography, and background aerosol profile. As the cloud layer near the Chilean coast is thinner than the cloud layer west of Mexico, the radiative forcings due to aerosol-radiation interactions for the reflecting aerosol are smaller for ChileROI than MexicoROI. This is because clouds reduce the amount of radiation reaching aerosol below, and because the radiative effect of reflecting aerosol above a reflecting surface is strongly reduced. Furthermore, the average solar insolation is stronger near the equator, which can contribute to the differing ERF_{NDG} values for these two ROIs. One explanation for differing ERF_{NDG} values is that in Chile the anthropogenic SO2 emissions are more pronounced when comparing to BC and OC than in Mexico. In addition, a large portion of the anthropogenic aerosol particles in MexicoROI are transported to altitudes above the optically thickest part of the cloud layer, making the RFARI for NO_BC more pronounced for MexicoROI. Previous studies have shown that low-level clouds are generally poorly represented by atmospheric general circulation models (Klein et al., 2013). It has to be noted that like many global climate models, ECHAM-HAMMOZ underestimates the persistent stratocumulus deck west of Chile (Stevens et al., 2013). It is therefore to be expected that the ERF values found here would be stronger if the Sc decks west of Chile and Mexico were simulated more realistically. Though, as Neubauer et al. (2019) showed, improving Sc representation in the models might not directly lead to larger ERF values.

For the MITIG simulation, the estimated ERF_{NDG} was positive for both ChileROI and MexicoROI. This demonstrates that the aerosol-cloud interactions and rapid adjustments due to BC and OC removal counter-act the negative RFARI caused by reduced BC emissions, leading to an overall warming effect. The reduction in emission strength of BC and OC in the MITIG scenario was based on simplified assumptions and the ERF_{NDG} values obtained here would probably change if a more realistic scenario was used.

Since one of our aims was to investigate how much emission location affects the obtained results, we performed another simulation where we removed all anthropogenic SO2 emissions from China. We found that the ERF efficiency, EFF_{ERF}, that is, the global ERF_{NDG} per emitted unit mass and time of SO2, for SO2 reductions in Chile and Mexico was 20 times higher than the efficiency calculated for Chinese SO2 emission reductions. We argued that the main cause of this difference is the non-linearity of aerosol-cloud interactions, which leads to a saturation of these effects over China, where anthropogenic emission strengths are amongst the highest of the world. Other possible causes include different insolation levels and differences in local meteorology.

In this study all simulations were nudged toward the same wind and surface pressure fields. This helped to greatly reduce modeled climate variability and thereby uncertainty intervals when comparing to similar studies without nudging (Baker et al., 2015; Cherian et al., 2017; Kühn et al., 2020). However, the uncertainty intervals obtained for the ERF_{NDG} values still were of the order of the ERF_{NDG} values themselves. This is partly due to the small area affected by the emissions studied here and partly due to further modeled climate variability which cannot be suppressed through nudging as performed in this study. A trade-off for the reduced uncertainty intervals due to nudging is the suppression of some aerosol-related rapid adjustments in the model. Simulations with free meteorology or even fully coupled Earth system models may therefore
find different values for the effective radiative forcings calculated here (Forster et al., 2016). Furthermore, TOA radiative forcing may not be the optimal measure for analyzing the warming potential of BC, as the relationship between BC climatic impacts and TOA forcing is complex (Flanner, 2013; Yang et al., 2019).

The ROI-averaged ERF\textsubscript{NO\_BC} values for Chile\textsubscript{ROI} and Mexico\textsubscript{ROI} for NO\_BC simulation were in the range of \(-0.2\) to \(0.3\) W m\(^{-2}\). Smith et al. (2020) estimated that the global ERF of anthropogenic greenhouse gases is 2.89 W m\(^{-2}\) between 1850 and 2014. Note that this global average is not directly comparable to the regional values we obtained here, because a global average of the radiative effects of BC mitigation in only Chile or Mexico would be much lower, while BC mitigation in other countries would most likely lead to different radiative forcings. Our findings support the general incentive that in some regions mitigating BC might help to reduce both regional and global warming on a short-term time scale, as an additional measure for CO\(_2\) reductions. However, this does not apply to all domains or nations, especially since the net effect of aerosol reductions is still highly uncertain on the global scale (Bellouin et al., 2020). The results obtained in this study underline how the changes in radiative balance due to aerosol mitigation are not linearly dependent on the total aerosol mass emitted. In other words, at some locations aerosol mitigation can reduce local warming or cooling effects more efficiently than in other regions, even if the emissions are relatively small. Besides climatic effects, SLCF mitigation could lead to improved surface air quality and thereby reduce negative health effects of these harmful pollutants. Thus, this may encourage also countries with fairly small yearly emissions to include SLCF mitigation in their climate and air quality efforts. In addition, limiting the region of analysis to areas with notable changes in aerosol and cloud properties provided a good platform to study the ERF for anthropogenic aerosol reduction, and thus this type of approach could be used to evaluate the effects of SLCF mitigation for other countries as well.

Conflict of Interest
The authors declare that they have no conflict of interest.

Data Availability Statement
The ECHAM6-HAMMOZ model is made available to the scientific community under the HAMMOZ Software License Agreement, which defines the conditions under which the model can be used. The license can be retrieved from https://redmine.hammoz.ethz.ch/attachments/291/License_ECHAM-HAMMOZ_June2012.pdf. The model data can be reproduced using ECHAM-HAMMOZ model revision 5914 from the repository https://redmine.hammoz.ethz.ch/projects/hammoz (HAMMOZ consortium, 2019). The settings for the simulations are given in the Fairdata Etsin service https://etsin.fairdata.fi/dataset/08a0ea09-19ea-4ce1-9283-cdb6568e7676. All emission input files, except ECLIPSE V6a are ECHAM-HAMMOZ standard and are available from the HAMMOZ repository (see https://redmine.hammoz.ethz.ch/projects/hammoz, HAMMOZ consortium, 2019).

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