ANTHROPOGENIC INFLUENCE ON CLIMATE THROUGH CHANGES IN AEROSOL EMISSIONS FROM AIR POLLUTION AND LAND USE CHANGE

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Anthropogenic influence on climate through changes in aerosol emissions from air pollution and land use change

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List of papers and author's contribution

**Paper I: Global emissions of terpenoid VOCs from terrestrial vegetation in the last millennium**


In this study I contributed with the design the model experiments, implemented and performed the model simulations for one of the two the models, analyzed the results from both models and wrote most parts of the manuscript.

**Paper II: Aerosol size distribution and radiative forcing response to anthropogenically driven historical changes in biogenic secondary organic aerosol formation**


In this study I contributed to the planning of the simulations, provided input data and helped implementing it in the chemical transport model. I took part in the discussions and analysis of the results and wrote the sections of the manuscript related to my work.

**Paper III: Amplification of Arctic warming by past air pollution reductions in Europe**


In this study I contributed with the design the model experiments, model setup, analyzed most of the results and wrote a large fraction of the manuscript.
Paper IV: Future response of temperature and precipitation to reduced aerosol emissions as compared with increased greenhouse gas concentrations


In this study I contributed with the design the model experiments, implemented and performed part of the simulations, analyzed the results and wrote most of the manuscript.
List of papers not included in the thesis


Abstract

Particulate matter suspended in air (i.e. aerosol particles) exerts a substantial influence on the climate of our planet and is responsible for causing severe public health problems in many regions across the globe. Human activities have altered the natural and anthropogenic emissions of aerosol particles through direct emissions or indirectly by modifying natural sources. The climate effects of the latter have been largely overlooked. Humans have dramatically altered the land surface of the planet causing changes in natural aerosol emissions from vegetated areas. Regulation on anthropogenic and natural aerosol emissions have the potential to affect the climate on regional to global scales. Furthermore, the regional climate effects of aerosol particles could potentially be very different than the ones caused by other climate forcers (e.g. well mixed greenhouse gases). The main objective of this work was to investigate the climatic effects of land use and air pollution via aerosol changes.

Using numerical model simulations it was found that land use changes in the past millennium have likely caused a positive radiative forcing via aerosol climate interactions. The forcing is an order of magnitude smaller and has an opposite sign than the radiative forcing caused by direct aerosol emissions changes from other human activities. The results also indicate that future reductions of fossil fuel aerosols via air quality regulations may lead to an additional warming of the planet by mid-21st century and could also cause an important Arctic amplification of the warming. In addition, the mean position of the intertropical convergence zone and the Asian monsoon appear to be sensitive to aerosol emission reductions from air quality regulations. For these reasons, climate mitigation policies should take into consideration aerosol air pollution, which has not received sufficient attention in the past.

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Abbreviations

BVOC: Biogenic volatile organic compound
SOA: Secondary organic aerosol
NorESM: Norwegian Earth system model
GEOS-Chem-TOMAS: Goddard Earth Observing System - Two-moment aerosol sectional
MEGAN: Model of emissions and gases and aerosols from nature
LPJ-GUESS: Lund–Potsdam–Jena general ecosystem generator
PM: Particulate matter
RF: Radiative forcing
RFP: Radiative flux perturbation
ARI: Aerosol-radiation interactions
ACI: Aerosol-cloud interactions
CLE: Current legislation emissions
MFR: Maximum technically feasible reductions
OC: Organic carbon
BC: Black carbon
TOA: Top-of Atmosphere
1. Introduction

1.1 Humanity: a geological force

We as a species have modified our surrounding natural environment for several millennia with the aim of generating more favorable conditions to survive and develop. Since the mid-twentieth century the pace of these transformations has accelerated exponentially causing environmental changes unprecedented in the last 420’000 years (Petit et al., 1998) and possibly pushing towards irreversible changes (Crutzen et al., 2000; Crutzen et al., 2002; Steffen et al., 2011). This has led scientists to compare humanity to an emerging geological force capable of causing global changes that were unthinkable two centuries ago (Crutzen et al., 2000; Crutzen et al., 2002; Steffen et al., 2011; Revkin, 1992). The human footprint on the environment is evident as ocean acidification, air and water pollution, changes in fresh water availability, land use change, loss of biodiversity, anthropogenic climate change, among others (Rockström et al., 2009; Steffen et al., 2015). Interestingly, many of these human-induced environmental changes are interconnected and can perturb the complex Earth system in non-trivial manners. Our urgent need to understand our planet is also a unique opportunity to learn about it more than ever before.

This thesis explores how two of these environmental problems: air pollution and land use change can influence another environmental problem: climatic change. The interconnecting agents between them in this work are atmospheric aerosols and their changes caused by human activities.

1.2 Atmospheric aerosol particles in the Earth system

Atmospheric aerosol particles are liquid or solid particles suspended in the air and are found throughout the atmosphere. They are generated naturally or by human activities. For example oceans, vegetated areas, wildfires and deserts emit natural aerosol precursors and aerosol particles to the atmosphere, while anthropogenic aerosols and precursors are generated from industrial activity, the transport sector, agriculture and electric power generation. Aerosol particles enter the atmosphere in a condensed phase (primary aerosol) or form by condensation of gaseous precursors (secondary aerosol). The parti-
Aerosol particles vary in shape, size (a few nm to over 100 µm) and chemical composition. The main chemical components include mineral compounds, sea salts, sulfates, nitrates and several different forms of carbonaceous compounds. After emission or formation in the atmosphere aerosol particles undergo several microphysical and chemical processes that can modify their properties. Aerosol particles are eventually removed from the air through gravitational settling, dry scavenging or precipitation processes. The typical atmospheric residence time of an aerosol particle is largely dependent on size and location, and varies between a few hours and a week or more. Their short residence time is the reason why higher concentrations of aerosols are typically found near the place of emission or formation.

Aerosol particles have an important effect on the Earth’s energy budget and hence the global climate. Depending on the size and chemical composition, they can serve as nuclei for cloud droplets and crystals. Therefore, changes in aerosol emissions and atmospheric loading can modify the optical and microphysical properties of clouds (Twomey 1977; Albrecht 1989). Additionally, aerosol particles absorb and/or scatter electromagnetic radiation. Changes in both natural and anthropogenic aerosol emissions have then the potential to modify the climate through Aerosol-Cloud-Interactions (ACI) and Aerosol-Radiation-Interactions (ARI).

Aerosol air pollution, also called particulate matter (PM) pollution, is one of the largest environmental problems. It causes about seven million premature deaths annually by increasing the likelihood of stroke, heart disease, lung cancer, and respiratory diseases (WHO, 2015). PM pollution is also responsible for the second largest climate forcing during industrial times. The net anthropogenic aerosol effect on climate is still uncertain, but has very likely had a net cooling effect (Boucher et al., 2013). Due to the adverse health effects of PM pollution, future policies will likely cause a reduction of emissions with associated climatic effects. The aerosol pollution – climate interactions become of strategic relevance once global warming limits and adverse health effects of air pollution are considered simultaneously.

Aerosol emissions are spatially inhomogeneous and changes in those emissions can drive regional effects on precipitation and surface temperature that differ from the effects caused by well-mixed greenhouse gases. For example, Arctic surface temperatures appear to be more sensitive to aerosol emission changes than to greenhouse gas emission changes (Najafi et al., 2015). Furthermore, interhemispheric differences in aerosol emissions are also thought to have an important effect on the mean position of the Inter-Tropical Convergence Zone (ITCZ) (Rotstayn and Lohmann 2002; Kristjansson et al. 2005) and the Asian monsoon system (Ramanathan et al. 2008). Once regional effects are brought into the picture, unanticipated aerosol effects
could take us by surprise if overlooked. A few studies have looked into the main climatic differences and similarities between aerosol and greenhouse gases (Xie et al., 2013; Wang et al., 2016), however to my knowledge no study has linked both anomalous atmospheric and oceanic energy transport to changes in the climatic patterns induced by aerosol emissions changes.

Additionally, the research on the human influence on aerosol concentrations has been mostly associated to changes in direct emission of anthropogenic aerosols. The natural aerosol emission changes caused by human perturbations on natural sources has been much less studied. Mankind, however, has modified about half of the land surface of the planet (Vitousek, 1997). Several million hectares of native forest are replaced each year with crops or pastures (FAO, State of the World’s Forests 2016, Chapter 2). This massive conversion of forested areas to agricultural areas has modified the fluxes of gaseous aerosol precursors to the atmosphere (Tanaka et al., 2012; Pacifico et al., 2012; Unger, 2014), and the atmospheric aerosol loading (Unger, 2014). Only Unger (2014) has quantified the historical land-use effects on climate through aerosol radiation interactions, however the climate effects through aerosol cloud interactions have not been targeted before.

1.3 Objectives

This thesis aims to quantify previously unexplored aspects of the human influence on climate via aerosols, mainly to help improve assessments and strategies on climate change. The aforementioned aspects are: land use radiative forcing and climatic responses to spatially heterogeneous aerosol forcing of past and future PM pollution, which are formulated as the following objectives.

Objective A: Historical land use - aerosol effects on the Earth’s energy budget. The first main objective of this thesis was to estimate the land use driven changes in natural aerosol emissions and the associated climate forcing caused by those changes in the past 1000 years. Secondary objectives within this main objective included:
(1) An estimate of the ACI land-use climate forcing, in addition to the ARI component.
(2) A comparison of the aerosol land-use climate forcing with the total anthropogenic aerosol climate forcing in the past. Papers I and II address objective A.

Objective B: Recent past and near future fossil fuel air pollution effects on surface temperature. The second main objective was to estimate the temperature effects of future fossil fuel aerosol emissions by 2050. This is
largely motivated by the apparent conflict between cleaner air and a warmer climate. The historical case of European sulfate emission reduction between 1980 and 2005 is studied and serves as an example that shows how deeply interconnected climate and air pollution are. Papers III and IV address objective B.

**Objective C: Regional climate change patterns driven by aerosol emission changes.** The third main objective of the thesis was to investigate beyond the radiative effects caused by regional aerosol emission changes by studying the associated full climatic responses. Central to the analysis is the understanding of meridional energy transport in ocean and atmosphere caused by localized aerosol Radiative Flux Perturbations (RFP) and how it differs from a response to more spatially homogeneous RFPs (i.e. from long-lived greenhouse gases). A secondary objective was to form a conceptual understanding of how RFP and ocean column heat uptake define the degree of meridional energy transport compensation between atmosphere and ocean. Papers III and IV address objective C.
2. Background

2.1 Natural aerosols from terrestrial vegetation

In addition to emitting primary biological particles, vegetated areas emit large amounts of trace gases known as Biogenic Volatile Organic Compounds (BVOCs) that form Secondary Organic Aerosol (SOA) after chemical processing in the air. There are thousands of types of BVOCs emitted by terrestrial vegetation into the atmosphere. However, isoprenoids (isoprene, monoterpenes and sesquiterpenes) and their oxidation products are abundant (over 50% of emitted BVOCs, Guenther et al. 2012) and have been recognized as important sources of SOA in different environments (Hallquist et al., 2009). Conservative estimates suggest that about 3%, 10% and 20% of the mass emitted as isoprene, monoterpenes and sesquiterpenes, respectively, form SOA (Pye et al., 2010), totaling to about 25 Tg/year. However, these numbers could be as high as 10% (isoprene), 20% (monoterpenes) and 40% (sesquiterpenes) yielding a total SOA production of 70 Tg/year. Of all vegetation types, forests are by far the largest sources of BVOCs on a global scale (Guenther et al., 1995, 2006). Additionally, a large fraction of the submicron dry aerosol mass over forested areas originates as SOA (Jimenez et al., 2009). This also means that aerosol particles formed as SOA typically dominate the total particle number concentration in clean forested environments and are very likely important components that define cloud properties in those environments. For this reason, this thesis is focused on ARI and ACI of changes in SOA rather than primary biological particles. The major driver of BVOC emission changes is very likely land use change, however BVOC emissions also depend on biotic and abiotic environmental conditions. Abiotic controllers include leaf temperature, photosynthetically available radiation, soil water content, carbon dioxide, and ozone concentrations.

2.2 Aerosols and air pollution

Historically, the largest global PM polluters have been the countries with developed industrial economies (Smith, 2011). However, strong regulations during the 1980s have led to a considerable reduction in PM pollution in those countries improving air quality significantly. Europe (including the former Soviet Union) has been the largest contributor to global reduction of
PM, followed by North America. Countries with developing economies, on the contrary, have seen a rapid increase in PM pollution levels in the last four or five decades driven by economic growth and low regulatory controls. Many of these countries (e.g. China, India) have large numbers of big urban centers with highly polluted air. Pushed by public health concerns, these countries are expected to eventually reverse their polluting emission trends to guarantee cleaner air to their citizens. However, the timing and the success of these policies is uncertain and very dependent on each country’s own efforts. Fig. 1 shows the future fossil fuel emission pathways in two scenarios (Klimont et al., 2016a,b) for three of the main PM polluters: Black carbon (BC), Organic Carbon (OC) and sulfur dioxide (SO₂). The Current Legislation Emission (CLE) scenario assumes present-day legislation in future emissions of pollutants, while the Maximum technically Feasible Reduction (MFR) emission scenario assumes a full implementation of present-day technology to control PM emissions in all nations. By 2030 there is a large difference between total emissions depending on which scenario is followed. Such difference in emissions is very likely equivalent to millions of lives saved every year.

Figure 1: Future emissions of SO₂, BC and OC under the MFR and CLE scenarios.
2.3 Aerosol effects on climate

Aerosols and clouds affect the energy balance of the planet, respectively they reflect 5.0 and 47.3 W/m² of the incoming solar radiation back to space (Yu et al., 2006, Boucher et al. 2013). Clouds also prevent 26.2 W/m² of thermal radiation emitted by the Earth’s surface and atmosphere from escaping to space (Boucher et al. 2013). Therefore clouds induce a net cooling of 21.1 W/m², which is about four times larger than the cooling caused by aerosol particles (Fig. 2). However, aerosol particles serve as seeds for cloud droplets and ice crystals. Changes in the aerosol properties can therefore modify the optical and physical properties of clouds and hence the energy balance of the planet (Twomey, 1977; Albrecht, 1989).

2.3.1 Radiative forcing definition and historical estimates

A Radiative Forcing (RF) is an instantaneous change in the radiation balance of the planet and it quantifies the immediate energetic perturbation on the system caused by any given agent. A Radiative Flux Perturbation (RFP) is similar to the RF, but includes the energetic responses or feedbacks after the RF is introduced. The RFP measures the energy imbalance of the planet more realistically than the RF since it is not restricted to the immediacy after the RF is introduced, but can be estimated at any time. Typically the RF is the metric used in most studies and is quantified at the tropopause or top-of-atmosphere (TOA). The best model estimates of the RF caused by human-induced ARI and ACI, during industrial times lie between +0.15 and -0.85 W/m², and 0.0 and -1.2 W/m², respectively (Boucher et al, 2013). Satellite-based estimates give a better constrained total anthropogenic aerosol (ARI+ACI) RF between -0.45 and -0.95 W/m² (Boucher et al, 2013). Fig. 2 summarizes the net global TOA radiative effects of aerosols and clouds, and the historical anthropogenic influence on aerosol-radiation and aerosol-cloud effects based on Boucher et al. (2013). The ARI RF caused by changes in SOA during industrial times through land use induced changes on BVOC emissions have only been estimated previously by Unger (2014), yielding a warming of +0.09 W/m², however the ACI RF has not been estimated before paper II.
2.3.2 Climate effects of a spatially inhomogeneous radiative forcing

An aerosol-induced RF is spatially uneven and may cause changes in surface temperature and precipitation that differ significantly from those caused by a well-mixed greenhouse gas RF (Rotstayn and Lohmann 2002; Kristjansson et al. 2005). Two distinct features caused by historical (20th century) changes in emissions of aerosol particles are present in observational and modelling studies: 1) A differential heating between hemispheres (Stern and Kaufmann, 1999 and 2000) due to differences in the location of aerosol emissions, loading and RF; and 2) A shift in precipitation patterns over tropical regions caused by a southward displacement of the mean position of the ITCZ induced by a reorganization of atmospheric circulation which adjusts to the differential heating of the hemispheres (Ridley et al. 2015, Baker et al., 2015).
2.3.3 Arctic climate effects of aerosol emissions

Climate models suggest that the increase of anthropogenic aerosol emissions masked 60% of the Arctic warming that would have resulted if only anthropogenic greenhouse gases had increased during the period 1913-2012 (Najafi et al., 2015). Globally, the equivalent number is approximately 30% (Storelvmo et al., 2016), meaning that Arctic surface temperatures have been about twice as sensitive to anthropogenic aerosol changes than global surface temperature in the recent past. In the last three to four decades, however, the Arctic has warmed at an accelerated pace, coinciding with strong global reductions of PM, led by Europe and North America. Few studies have studied the possible role of PM reduction in the last decades and the strong observed Arctic amplification. For example, Yang et al. (2014) studied the combined effect of increased Asian PM pollution with reduced North American and European PM pollution, but no study before paper III had studied the effect from individual regions on recent Arctic warming. A strong Arctic amplification is undesirable since it can increase the likelihood of triggering abrupt climatic changes via non-linear dynamic mechanisms in the Earth system (or tipping elements) such as the collapse of the Greenland ice sheet, greenhouse gas release from thawing permafrost or altering the Atlantic meridional overturning circulation (Lenton et al 2008). Hence, building a better understanding of the drivers of Arctic climate change is important because of the possible impact on the rest of the planet (Alley et al., 2002; Cohen et al. 2014).

2.4 Meridional energy transport in the climate system

Equation 1 expresses the conservation of energy for latitude bands from the bottom of the ocean to the top of atmosphere with infinitesimal meridional thickness and assumes that all energy gained (lost) by the system at the TOA goes to (comes from) the oceans.

\[
\Delta \frac{\partial O(y)}{\partial t} + \Delta \frac{\partial (F_O(y) + F_A(y))}{\partial y} = \Delta Q_{TOA}(y)
\]

In Eq. 1 \( \Delta \) is the difference between a pair of scenarios or different periods, \( O(y) \) is the column-integrated, area-averaged and zonally-integrated oceanic energy content per unit length, \( Q_{TOA}(y) \) is the area-averaged zonally-integrated net downward flux at the TOA, and \( F_O(y) \) and \( F_A(y) \) are the meridional energy transport in the ocean and atmosphere, respectively. Equation 1 is used to explore conceptually the degree of compensation in meridional transport by atmosphere and ocean caused by a RFP. Equation 1 implic-
itly states the conditions for Bjerknes compensation at a given latitude ($\phi = y/a$, $a$ being the Earth’s mean radius), that is: $\Delta \partial O(y)/\partial t = \Delta Q_{\text{TOA}}(y)$.

Studies of changes in the meridional heat transport of ocean and atmosphere have typically focused on either greenhouse forcing or large freshwater inputs in northern hemisphere (NH) high-latitudes (Shaffrey et al., 2006, Farneti and Vallis, 2013). A large degree of compensation of ocean and atmosphere heat transport is found (Bjerknes compensation, Bjerknes 1964), particularly at mid-latitudes. However, ocean-atmosphere meridional heat transport changes caused by anthropogenic aerosol RF have not been studied before paper IV. Additionally, most studies aimed to quantify and understand Bjerknes compensation either on decadal variability or centennial to millennial variability. Multidecadal changes of the Bjerknes compensation are also important from a societal perspective due to the typical timescales that societies need to adapt to the associated climatic responses caused by oceanic and atmospheric circulation changes. However, little attention has been devoted to study Bjerknes compensation on such timescales. In paper IV we compare the compensation of a localized aerosol RFP and a more globally homogeneous well-mixed greenhouse gas RFP.
3. Methodology

Two modelling approaches were used in this work. The first approach is based on a chemical transport model forced with changing BVOC emissions from two land emission models to study the radiative effects via SOA changes. The main advantage of this modelling approach is that it uses detailed aerosol dynamical and aerosol chemical processes describing SOA formation from BVOC oxidation. However, the chemical transport model is forced with prescribed dynamics (meteorology) neglecting aerosol-cloud-climate feedbacks (See e.g. Boucher et al., 2013). This method can give an estimate of the direct radiative and the cloud albedo effect of aerosols. A disadvantage of this method is that it assumes fixed vertical wind velocity to calculate the maximum water vapor supersaturation that drives cloud droplet activation.

The second approach is based on earth system model results. The main advantage of this method is that the radiative effects of aerosol changes feedback on clouds and climate that can further affect the aerosol properties. This method is not limited to radiative forcing estimates, but allows for a full estimate of climate effects (e.g. precipitation, surface temperature). These models explicitly solve the equations of motion in the atmosphere and ocean, as opposed to the chemical transport model. In addition to the direct radiative and the cloud albedo effect of aerosols it also estimates the cloud lifetime and the semi-direct effect of aerosols (Boucher et al., 2013).

Both approaches have disadvantages in terms of limited aerosol and cloud lifecycle treatment, but maybe more importantly, due to model inability to resolve the scales at which aerosol and cloud processes operate. These processes are parametrized in large scale models, introducing uncertainty.

3.1 Chemical transport and surface emission modelling

The objective A, addressed by papers I and II, builds upon the work presented in my licentiate thesis (Acosta Navarro 2015). On Paper I, the BVOC emission models MEGAN and LPJ-GUESS were driven by climatic and land-use conditions from the past millennium to study environmental effects on isoprene, monoterpane and sesquiterpene emissions. The models use
standard emission factors representing different vegetation types. Land use changes follow from anthropogenic crops and pasture reconstruction that change with time and replace natural vegetation species. Solar radiation, soil moisture, leaf temperature and leaf area index affect the emission magnitude based on empirical factors that multiply the standard emission factor. Carbon dioxide concentrations were allowed to inhibit only isoprene emissions in both models.

Using isoprene, monoterpene and sesquiterpene emissions from paper I, a chemical transport model (GEOS-Chem-TOMAS) was used to estimate oxidation of emitted BVOCs, their SOA yield, and the associated RF via ARI and ACI. Forcing the same meteorology on each run allowed estimating the RF. Aerosol size distribution in the model is represented by 15 bins ranging from 3nm to 10μm. Aerosol and gas emissions except BVOCs were prescribed (van Donkelaar et al. 2008) while oxidants were fixed at present-day. BVOC emissions representing the beginning and end of past millennium were the only variables that changed between different pairs of model simulations (more details are found in Paper II). BVOC oxidation products form non-volatile SOA at fixed yields (Pye et al., 2010). Particles undergo kinetic, diffusion-limited growth where the gas condensation is proportional to the aerosol surface area. Schematic 1 displays the model configuration to address objective A.
3.2 Earth system modelling

The main tool for addressing objectives B and C is the Norwegian Earth system model (NorESM1, Bentsen et al. 2012, Iversen et al. 2012). NorESM1 is a numerical model built of individual, but interactive land, atmosphere, ocean, and sea ice models (See Schematic 2). The atmospheric component contains an explicit description of the aerosol lifecycle of dust, sea salt, sulfate, black and organic carbon particles, which makes it suitable for studies focusing on aerosol-climate interactions (Kirkevåg et al., 2012). Aerosols can be internally or externally mixed, hygroscopic growth and particle absorption or reflection of radiation are taken into account. Additionally, activation of aerosols to cloud droplets follows Köhler theory (Abdul Razzaq and Ghan, 2001) for further cloud droplet radiation calculations. In summary: ARI and ACI (warm clouds) interactions are both accounted for in NorESM1.

Paper III includes a total of eighteen simulations representing two different scenarios that span the period from 1980 to 2005. Aerosol and gaseous aerosol precursor emissions, as well as greenhouse gas concentrations follow CMIP5 historical estimates (Lamarque et al., 2010) driving the climate in the control scenario. In the perturbed scenario, anthropogenic sulfate aerosol and precursor emissions over Europe are fixed at the 1980 level and all other conditions are exactly the same as in the control experiments. Due to the large Arctic climate natural variability and the short temporal span of the simulations (1980-2005), each simulation is repeated nine times for each scenario to distinguish the signal from the noise. Both ensemble simulations (control minus perturbed) are compared to single out the climate effects of declining sulfate aerosols over the European continent.

In paper IV a total of twelve simulations are included representing four future scenarios (three simulations per scenario). Three fossil fuel aerosol emission scenarios (CLE, MFR and 2005) are followed (Klimont et al. 2016) simultaneously with greenhouse gas emissions from RCP4.5 (Taylor et al., 2012). The fourth scenario used fixed 2005 greenhouse gases and CLE emissions. The CLE scenario is built upon the assumption of present-day emissions controls and future economic growth of each country. As a result of this, most developed nations continue reducing their emissions targeting air pollution, while some developing nations (China) reach a peak in emissions during the 2020s and others (India) increase their emissions continuously. In the MFR scenario, all nations reduce fossil fuel sulfate, black and organic carbon emissions according to the maximum technically feasible reductions by 2030 (See table 1 in paper IV for more details on the simulations).
Schematic 2: The Norwegian Earth system model NorESM1.
4. Results and discussion

4.1 Historical land use - aerosol effects on the Earth’s energy budget.

Between the beginning and the end of the past millennium there was an increase of 115% in the area dedicated to agriculture (Kaplan at al., 2011). A significant fraction of the increase in agricultural land happened at the expense of natural vegetation which decreased by 17% during the same period. The largest part of these changes happened after the industrialization: the increase in agricultural land from the beginning of the nineteenth century until the end of the twentieth century was 74%, while the decrease in natural vegetation was 14%. Carbon dioxide and mean global surface temperature increased by about 21% and 0.8 K, respectively from the beginning of the nineteenth century until the end of the twentieth century. Both models MEGAN and LPJ-GUESS (paper I) indicate that global isoprene emissions decreased by about 20% between the beginning and end of the past millennium driven by land-use changes and emission inhibition from carbon dioxide concentrations, while mono- and sesquiterpenes either increased or decreased slightly during the same time period and were controlled mostly by surface air temperature changes. Despite the agreement between MEGAN and LPJ-GUESS, the total isoprene emission magnitude and the driver of changes are different. In MEGAN land-use changes cause most of the isoprene emission reductions, while in LPJ-GUESS carbon dioxide emission inhibition is the main cause for isoprene emission reductions on a planetary scale. However, both models predict consistent reductions in BVOC emissions in regions with large deforestation and this seems to be the most robust feature.

The large decrease in isoprene emissions in both MEGAN and LPJ-GUESS causes a decrease in climatically relevant aerosol particle concentrations leading to an ARI RF between +0.02 and +0.16 W/m² (Paper II) and compares well with Unger (2014) estimate of +0.09 W/m². The results are not directly comparable due to the fact that Unger (2014) considered only land-use change as the driver of emissions changes, while we additionally considered temperature, solar radiation, soil moisture and carbon dioxide concentrations. The ACI RF falls between -0.01 and -0.06 W/m², cancelling partially the ARI RF. The combined effect falls between +0.02 and +0.12 W/m², still causing a warming effect (Fig. 3). Low- and mid-latitude regions with
large deforestation show the largest positive RF (<0.5 W/m²), caused by the large BVOC flux changes in those regions, confirming that land use change is the dominant driver. Globally averaged, the BVOC-SOA effects on climate are about an order of magnitude lower than the global aerosol RF from fossil fuel and biomass burning in the industrial era and have an opposite sign. However, in regions with little air pollution from fossil fuel or biomass burning, but substantial land-use changes, the anthropogenic aerosol RF could be dominated by changes in secondary organic aerosol.

![Figure 3: Annual mean change from year 1000 to year 2000 in best estimates of the combined radiative effect with MEGAN BVOC emissions and anthropogenic emissions off (left), and the combined radiative effect with LPJ-GUESS BVOC emissions and anthropogenic emissions off (right). Global mean changes are +0.05 and +0.02 W/m², respectively. Figure taken from D’Andrea et al., (2015).](image)

4.2 Recent past and near future fossil fuel air pollution effects on surface temperature.

There has been a 70 to 80% reduction in anthropogenic SO₂ and SO₄ emissions over Europe between 1980 and 2005 (Smith et al., 2011). These reductions cause an increase in the global mean annual surface temperature of about 0.06 K, and of 0.5 K in the Arctic (paper III). The anthropogenic aerosol effects are further investigated in the future scenarios CLE and MFR (paper IV). The CLE scenario following current legislation policies predict a moderate global reduction (5-20%) of SO₂, OC and BC by 2030, while the MFR scenario predicts a strong reduction of 70-80% of the same pollutants (Fig. 1). Compared with a simulation in which aerosol emissions are fixed at the levels of 2005 (Fig. 4, purple line), the surface temperature response to the CLE aerosol scenario (Fig. 4, green line) by the 2040s is negligible, while there is an increase of 0.3 K caused by aerosol reductions in MFR (Fig. 4, blue line vs. purple line). Experiments with the same model indicate that a greenhouse gas (GHG) effect following the RCP4.5 scenario (Fig. 4,
green) would give a warming of 0.5 K in the 2040s as compared with a simulation with fixed greenhouse gas concentrations at the levels of 2005 (Fig. 4, red line). The Arctic warming by 2040s is 0.9 K if only aerosol emissions change following MFR and by 1.3 K if only greenhouse gases change following RCP4.5.

![Graph showing surface temperature anomalies](image)

**Figure 4:** Anomalies of the mean global surface temperature with respect to 2001-2010 values. The orange line displays observations from GISTEMP (Hansen et al. 2010). All dashed series represent means from individual runs and the thick solid lines are the ensemble means. All series are monthly means filtered with a 12-month running mean. Figure taken from Acosta Navarro et al. (2016b).

4.3 Regional climate change patterns driven by aerosol emission changes and the links to oceanic and atmospheric energy transport.

The reduction in sulfur-containing compounds over Europe has led to lower aerosol concentrations causing a reversal of the negative RFP induced by these particles before 1980. In other words part of the sulfate aerosol mask over the region has been removed allowing more radiation to be absorbed by the climate system (Fig. 5a). The positive RFP since the 1980s is expected because sulfate aerosols scatter solar radiation effectively and due to their
high hygroscopicity are efficient cloud condensation nuclei affecting cloud albedo and lifetime. Observations show an important increase in solar radiation at the surface over the European continent since the 1980s, which is very likely caused by the reduction in emissions of PM pollutants (Allan 2013 JGR). NorESM1 simulates a similar increase in surface solar radiation during 1980-2005 driven mostly by the SO2 reduction.

Coincidentally, the Arctic region has experienced a rapid warming during the last four decades. We show a remote and local influence of the sulfate aerosol reductions in Europe on Arctic climate (paper III). The model simulations indicate that a lower transport of aerosol particles together with an anomalously higher oceanic (+ 0.48 W/m²) and summertime atmospheric energy transport (+0.25 W/m²) from mid-latitudes to Arctic latitudes are likely responsible for about a third of the warming observed during the period 1980-2005. In addition, Arctic climate feedbacks have possibly reinforced the warming through lower surface albedo, higher water vapor concentrations, larger heat fluxes from the ocean to atmosphere, lapse rate and Plank surface temperature amplification, especially in the winter half of the year (Serreze and Francis, 2006; Graversen and Wang, 2009; Pithan and Mauritsen, 2014).

Figure 5: Annual mean (1996–2005) anomaly between historical and fixed European SO2 and SO4 emission simulations for the net (incoming minus outgoing) shortwave (SW) and longwave (LW) radiative flux at TOA (left), and the surface temperature (TS) (right). Stippling indicates statistical significance at 95% using a two-tailed Student’s t-test. Figure taken from Acosta Navarro & Varma et al. (2016a).

The strong removal of aerosol particles in MFR (paper IV) causes a clear interhemispheric difference in the RFP and the associated surface temperature change in the last 25 years of simulations (2024-2049). The TOA energetic imbalance is mostly located in the NH mid-latitudes causing remote effects in high and low latitudes. The Arctic region experiences a three times stronger warming than the globe, and receives more precipitation, while the annual, and zonal maximum of tropical rainfall is shifted northwards. The East Asian monsoon precipitation is stronger in MFR possibly due to an
intensification of the monsoon circulation. The more moderate reduction of aerosol emissions in CLE also causes a marked northward shift of the mean position of the intertropical convergence zone, however most of the statistically significant changes in precipitation happen over oceans. The well mixed greenhouse gas induced warming (GHG) shows more interhemispheric symmetry than the aerosol induced warming with a lower ratio of Arctic to global surface temperature change. In both MFR and CLE scenarios, there are considerable changes in the mean annual Hadley circulation as a result of the interhemispheric difference in RFP. These changes are larger than the ones caused by greenhouse gas increases. Compared to 2005 aerosol emissions, in both MFR and CLE scenarios, the atmosphere adjusts itself so it can transfer energy from the northern to the southern hemisphere (SH) (Figs 6a and b) and the northward shift of the intertropical convergence zone is a manifestation of that. In contrast, the greenhouse gas increase (RCP4.5) does not lead to an interhemispheric energy transport in the atmosphere (Fig. 6c) compared to fixed 2005 greenhouse gases. In general, there is a compensation between oceanic and atmospheric energy transport in both CLE and GHG at most latitudes (Figs. 6b and c). This is not seen in MFR where ocean and atmosphere transport energy to the SH (Fig. 6a). Additionally, in MFR there is a positive anomalous transport of water vapor (latent heat) to the Arctic region which causes part of the increase in rainfall and possibly enhances the amplification of the warming in that scenario.

Figure 6: Mean 2025-2049 zonally averaged meridional energy transport change for the atmosphere (\(F_A\)), ocean (\(F_o\)), and climate system (\(F_o+F_A\)) in the MFR (top left), the CLE (top right), and the GHG (bottom left) pairs of simulations. The x-axis shows latitude as \(\sin(\text{latitude})\) to display the relative area between parallel lines. Figure taken from Acosta Navarro et al. (2016b).
Figure 7 shows a schematic representation of the meridional energy transport in the climate system as a response to a RFP and illustrates two extreme cases with conditions where there should and should not exist Bjerknes compensation according to Eq. 1. Figure 7a indicates that if the meridional distribution of a RFP (energy gain) matches exactly the zonally averaged distribution of oceanic column heat uptake, there must exist Bjerknes compensation as a result of energy conservation. However, if the RFP is localized at certain latitudes but the ocean heat uptake is distributed homogeneously at all latitudes, there cannot exist Bjerknes compensation because it would violate energy conservation principles (Fig. 7b). The zonal ocean heat uptake distribution is therefore as critical as the RFP. In the CLE and GHG cases (Figs. 6b and c), where the TOA imbalance (RFP) and its meridional gradient are generally smaller than in MFR, the oceanic transport compensates most of the atmospheric response, as opposed to MFR where the oceanic energy transport reinforces the atmospheric energy transport at most latitudes and Bjerknes compensation is not maintained (Fig. 6a).

To summarize, the degree of meridional compensation between ocean and atmosphere is defined by the meridional anomalies of the TOA RFP and ocean column heat uptake. On decadal temporal scales, the model simulations show an ocean-atmosphere coupling mechanism that tends to redistribute energy across the Equator without Bjerknes compensation once a large aerosol RFP acts predominantly at mid-latitudes of one hemisphere, while the ocean column heat uptake is more evenly distributed in both hemispheres.

Figure 7: Left: Bjerknes compensation between oceanic ($F_o$) and atmospheric ($F_a$) meridional energy transport under identical net zonal mean of top of atmosphere RFP ($\Delta Q_{TOA}$) and zonal mean of ocean heat uptake ($\Delta \partial O/\partial t$). Right: No Bjerknes compensation under different $\Delta Q_{TOA}$ and $\Delta \partial O/\partial t$. The changes in $F_o$ and $F_a$ were chosen arbitrarily to illustrate the compensation effects. The only restriction is that their sum must be equal to the implied net change in $F_o+F_a$. 
5. Conclusions and outlook

We found that the human influence on the Earth’s energy budget (i.e. the radiative forcing) through secondary organic aerosol changes from land use in the past millennium is about an order of magnitude smaller than the influence caused by aerosol emissions from industry, energy, transport and biomass burning. Furthermore, the land use aerosol radiative forcing is strongest in the tropics and the southern hemisphere, where large deforestation has taken place. The uncertainty in our estimates is still considerable because many biological, chemical and physical processes are parametrized and/or incomplete in models. Better process understanding of plant production and emission of BVOCs, gas to particle conversion of oxidized BVOCs, aerosol removal and cloud micro- and macrophysics will probably lead to more accurate estimates in the future. Healthy forests are part of complex ecosystems that provide us invaluable services. Among others, forests buffer precipitation and surface temperature extremes, they currently take up excess carbon dioxide from the atmosphere (FAO, State of the world’s forests 2016) and help maintain precipitation downwind (Spracklen et al., 2012). Attempts to estimate climate effects exclusively via radiative forcing caused by land use changes might fall short. For this reason future efforts would greatly benefit from approaches linking vegetation changes to biophysical and biochemical feedbacks in a more comprehensive framework. Strategies aiming to mitigate climate change through land use management should also focus on the several other climate services they provide.

A strong northern hemisphere aerosol reduction following stringent air pollution controls (MFR scenario) could lead to an additional global warming of about 0.3 K by 2050. This reveals an apparent conflict between air quality and climate change policies. The climate effects caused by past emission reductions in Europe (and possibly North America) highlight the same conflict, especially when it comes to Arctic warming. However, the reduction of air pollution in the other regions of the planet is an imperative due to its immediate effects on human health. Large cities, where most of the global population will live by 2050, will face dire conditions in the future unless effective measures are taken to clean up the air. A contribution to global warming of 0.3 K is not negligible, but it is still small compared to the greenhouse induced warming projected by 2100. Our findings simply reinforce any argument in favor of curbing carbon dioxide emissions as soon as
possible to successfully limit global warming and the associated myriad of socioeconomic consequences. These results further indicate that policies aiming to control climate change exclusively based on greenhouse gas emissions might not be enough if air quality is neglected.

The aerosol reduction following the MFR scenario could cause amplified Arctic warming, a northward shift of the ITCZ of about one latitudinal degree and modify the meridional energy transport in the ocean and atmosphere by a few percent due to changes in large scale motions. Even regional redistribution of aerosol emissions (CLE) can cause significant changes in atmospheric circulation and an ITCZ migration comparable to that of the MFR scenario. The meridional atmospheric energy transport in the tropics is tightly related to the Hadley circulation and the position of the ITCZ, while the meridional oceanic energy transport is tightly linked to the thermohaline circulation of the Atlantic Ocean. Future changes in the ITCZ position and the strength of the thermohaline circulation of the Atlantic Ocean would very likely have large societal implications in many regions of the planet (e.g. changes in North Atlantic climate state and extreme weather patterns, changes in the transient climate sensitivity, tropical rainfall variability). Furthermore, global climate models differ significantly in the magnitude and future changes of the Atlantic meridional overturning circulation which is responsible for shaping much of the NH Atlantic climate. There has been continuous monitoring of atmospheric circulation and the top-of-atmosphere energy budgets for at least thirty years, but the ocean heat budget, and especially oceanic circulation are still less well monitored. The ocean is a component of the climate system associated with slow feedbacks and irreversible climate changes that also deserves continuous monitoring.

Our results are based on numerical models. We acknowledge that there is still a large discrepancy in model estimates of aerosol effects on climate (IPCC Boucher et al., 2013), particularly the ones related to aerosol-cloud interactions. Multi-model simulations following the same scenarios can help understand that discrepancy, but only accurate representations of aerosol-cloud interactions in climate models systematically evaluated with observations can help reduce any underlying uncertainty. There has been a large effort in the last decades to understand aerosol processes, while an integral understanding of cloud processes has been less targeted. Advancing our cloud process understanding in parallel with aerosol process understanding should be a priority in the climate research community to successfully reduce the uncertainty associated to aerosol-cloud interactions. To quote Gentleman (2015), we should be “putting the clouds back in aerosol-cloud interactions”.

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This work illustrates how different components of the Earth system function in unison. The environment is likely linked in unexpected ways. Finding these links in the future will open many new and exciting trails.
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