Aerosol-Cloud-Precipitation Interactions - Studied using combinations of remote sensing and in-situ data

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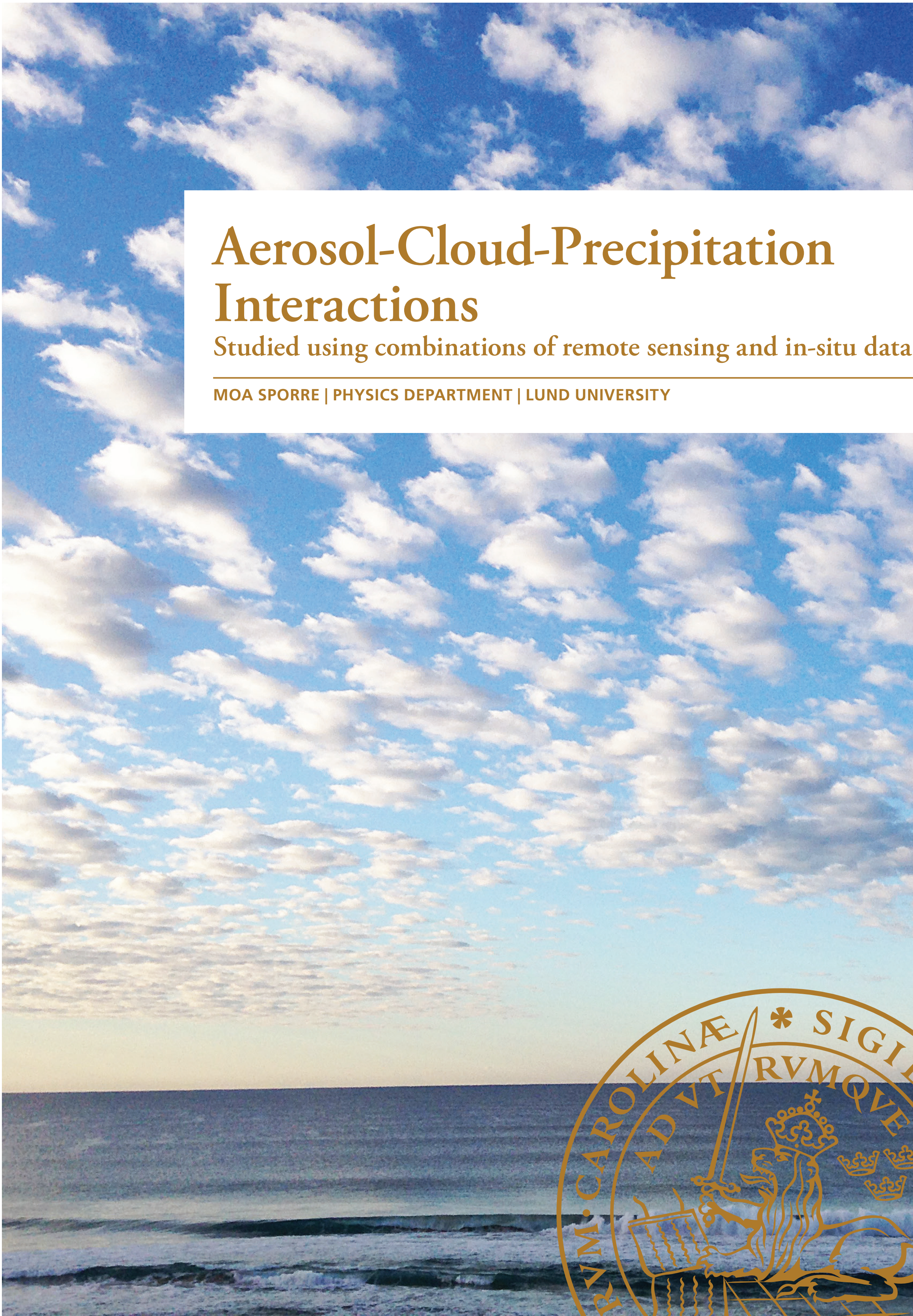

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MOA SPORRE | PHYSICS DEPARTMENT | LUND UNIVERSITY

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Moa Sporre

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<tr><td colspan="4">Abstract
Cloud droplets never form in the atmosphere without a seed in the form of an aerosol particle. Changes in number concentrations of aerosol particles in the atmosphere can therefore affect the number of droplets in a cloud. Higher concentrations of aerosol particles in the atmosphere lead to clouds with more droplets and if the amount of liquid water in the clouds stay the same, the droplets become smaller. Clouds with more, smaller droplets reflect more sunlight and may take longer to produce precipitation. In the research presented in this thesis, satellite data of clouds are combined with a range of other datasets to investigate how sensitive the cloud properties are to changes in the concentration of aerosol particles.

Cloud droplets were found to be smaller in low-level clouds formed in air with higher aerosol number concentrations over the ocean north of Scandinavia. This was also true for low-level and convective clouds over land in Sweden and Finland. The results regarding cloud optical thickness (COT), which is a measure of how much light a cloud reflects, was not as conclusive. For the low-level clouds over the ocean, the COT was higher in air masses with higher aerosol number concentrations. Differences in meteorological conditions in the clean and polluted air masses may however explain some of the differences in COT. The low-level and convective clouds over land did not show any significant changes in COT with varying aerosol number concentrations. This may be caused by changes in cloud dynamics due to the smaller droplets in the clouds. Hence, the indirect aerosol effect could not be observed for clouds studied over land.

The precipitation intensity from the clouds over land and how this varied with changing aerosol loading was also investigated. For both low-level and convective clouds, the precipitation was found to decrease somewhat with increasing aerosol number concentrations. However, for the convective clouds, this relationship only appeared when the clouds were sorted according to vertical extent, as higher convective clouds tend to produce heavier precipitation.

How cirrus clouds at midlatitudes in the northern hemisphere are affected by the mass concentration of particulate sulphate present in the lowermost stratosphere (LMS) was investigated using satellite data. Changes in the LMS particle levels were caused by explosive volcanos that emit gases and particles into the stratosphere. Due to subsidence in the stratosphere at midlatitudes, the volcanic sulphate eventually enters the upper troposphere, increasing its sulphate concentration. The reflectance of the cirrus clouds decreased when there were more sulphate particles present in the LMS. Cirrus clouds warm the climate and a decrease in their reflectance hence cools the climate.</td></tr>
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Contents

Papers included in thesis

I. **Sporre, M. K.**, O'Connor, E. J., Håkansson, N., Thoss, A., Swietlicki, E. and Petäjä, T. Comparison of MODIS and VIIRS cloud properties with ARM ground-based observations over Finland. Manuscript in preparation.

II. **Sporre, M. K.**, Glantz, P., Tunved, P., Swietlicki, E., Kulmala, M. and Lihavainen, H., 2012. A study of the indirect aerosol effect on subarctic marine liquid low-level clouds using MODIS cloud data and ground-based aerosol measurements, *Atmospheric Research*, 116, 56-66. doi: 10.1016/j.atmosres.2011.09.014 (Reprinted with permission from Elsevier.)

III. **Sporre, M. K.**, Swietlicki, E., Glantz, P. and Kulmala, M., 2014. Aerosol indirect effects on continental low-level clouds over Sweden and Finland, *Atmospheric Chemistry and Physics*, 14, 12167-12179, doi:10.5194/acp-14-12167-2014.

IV. **Sporre, M. K.**, Swietlicki, E., Glantz, P. and Kulmala, M., 2014. A long-term satellite study of aerosol effects on convective clouds in Nordic background air, *Atmospheric Chemistry and Physics*, 14, 2203-2217, doi:10.5194/acp-14-2203-2014.

V. Friberg, J., Martinsson, B. G., **Sporre, M. K.**, Andersson, S. M., Brenninkmeijer, C. A. M., Hermann, M., van Velthoven, P. F.J.. and Zahn, A., 2015. Influence of volcanic eruptions on midlatitude upper tropospheric aerosol and consequences for cirrus clouds, *Earth and Space Science*, 2, 7, 285-300, doi:10.1002/2015EA000110.

The author’s contributions to the papers

I. I came up with the idea to compare the satellite and ground-based measurements. I obtained the MODIS satellite data and carried out the data analysis for the whole dataset. I wrote the majority of the manuscript.

II. I carried out the manual selection of the satellite data, wrote the code for the analysis programs, combined the different datasets and evaluated the data. I also wrote the paper.

III. I came up with the research idea, carried out the selection of the satellite data and obtained the other datasets. I also processed and evaluated the data and wrote the paper.

IV. I developed the research idea, selected the satellite data, created the program to obtain vertical profiles, combined the different datasets and analysed the data. I also wrote the paper.

V. I obtained and evaluated the satellite data. I was involved in the evaluation of the results regarding cirrus clouds and I wrote the parts of the paper regarding cirrus clouds.

Related publications

Peer reviewed papers

Fors, E. O., Swietlicki, E., Svenningsson, B., Kristensson, A., Frank, G. P. and **Sporre, M.**, 2011. Hygroscopic properties of the ambient aerosol in southern Sweden – a two year study. *Atmospheric Chemistry and Physics*, 11, 8343-8361.

Rissler, J., Nordin, E. Z., Eriksson, A. C., Nilsson, P. T., Frosch, M., **Sporre, M. K.**, Wierzbicka, A., Svenningsson, B., Löndahl, J., Messing, M. E., Sjogren, S., Hemmingsen, J. G., Loft, S., Pagels, J. H. and Swietlicki, E., 2014. Effective density and mixing state of aerosol particles in a near-traffic urban environment, *Environmental Science & Technology*, 48, 6300-6308, doi:10.1021/es5000353.

Kecorius, S., Kivekäs, N., Kristensson, A., Tuch, T., Covert, D. S., Birmili, W., Lihavainen, H., Hyvärinen, A.-P., Martinsson, J., **Sporre, M. K.**, Swietlicki, E., Wiedensohler, A. and Ulevicius, V., 2016. Significant increase of aerosol number concentrations in air masses crossing a densely trafficked sea area, *Oceanologia*. 58, 1-12, doi:10.1016/j.oceano.2015.08.001

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Conference abstracts as lead author

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Sporre, M. K., Swietlicki, E., and Glantz, P. A long-term study of the aerosol second indirect effect on convective clouds over southern Sweden. International Aerosol Conference 2010.

Sporre, M. K., Swietlicki, E., Glantz, P. and Kulmala, M. A long-term study of the aerosol effects on convective clouds over southern Sweden and Finland. European Geosciences Union General Assembly 2011.

Sporre, M. K., Swietlicki, E., Glantz, P. and Kulmala, M. A long-term study of the aerosol effects on convective clouds over southern Sweden and Finland. European Aerosol Conference 2011.

Sporre, M. K., Swietlicki, E., Glantz, P. and Kulmala, M. A study of how aerosols affect low-level clouds over the Nordic Countries using MODIS, ground-based, ECMWF and weather radar data. European Geosciences Union General Assembly 2012.

Sporre, M. K., Berghof, M. I. A., Frank, G., Martinsson, B. G. and Swietlicki, E. A comparison of cloud microphysical parameters between MODIS satellite data and ground-based measurements with the DAA. Nordic Aerosol Symposium 2014.

Abbreviations and symbols

α	proxy of cloud condensation nuclei concentration
ACI	aerosol cloud interaction relationship
AMF2	ARM mobile facility 2
ARM	Atmospheric Radiation Measurement program - US Department of Energy
BAECC	Biogenic Aerosols – Effects on Clouds and Climate
BALTEX	The Baltic Sea Experiment
BT	brightness temperature
CARIBIC	Civil Aircraft for Regular Investigation of the atmosphere Based on an Instrument Container
CCN	cloud condensation nuclei
COT	cloud optical thickness
CR	cirrus reflectance
CTH	cloud top height
CTP	cloud top pressure
CTT	cloud top temperature
CAPE	convective available potential energy
dbzc	average radar reflectance
DMPS	differential mobility particle sizer
dT	temperature difference between cloud base and cloud top
ECMWF	European Centre for Medium-Range Weather Forecasts
FMI	Finish Meteorological Institute
GDAS	Global Data Assimilation System
HYSPLIT	Hybrid Single-Particle Lagrangian Integrated Trajectory

IBA	ion beam analysis
IN	ice nuclei
IRW	infrared window approach
LMS	lowermost stratosphere
LWP	liquid water path
LTSS	lower-tropospheric static stability
N_x	number concentrations of aerosol particles with a diameter greater than x nm.
N_d	droplet number concentration
NCEP	The US National Centers for Environmental Prediction
MODIS	Moderate Resolution Imaging Spectroradiometer
MWR	microwave radiometer
PIXE	particle induced X-ray emission
PESA	particle elastic scattering analysis
PPS	Polar Platform System
PV	potential vorticity
RF	radiative forcing
r_e	cloud droplet effective radius
RH	relative humidity
S	sulphate mass concentration
SH	specific humidity
SMHI	Swedish Meteorological and Hydrological Institute
T_B	cloud base temperature
UT	upper troposphere
VIIRS	Visible Infrared Imaging Radiometer Suite
VOC	volatile organic compounds
w	vertical velocity

Populärvetenskaplig sammanfattning

Den här avhandlingen handlar om hur moln påverkas av luftburna partiklar, så kallade aerosolpartiklar. Moln bildas i atmosfären när vatten övergår från gasfas till vätskefas, det vill säga när vattenånga kondenserar. För att molndroppar ska kunna bildas krävs dock aerosolpartiklar, som vattenångan kan kondensera på. Antalet partiklar i luften kan därigenom påverka hur många molndroppar som bildas i ett moln.

När vi släpper ut avgaser, från till exempel bilar, släpper vi ut växthusgaser men även aerosolpartiklar i luften. Sedan den industriella revolutionen har vi således ökat antalet partiklar i luften, vilket i sin tur leder till att det kan bildas fler molndroppar i molnen. Om man har en bestämd mängd vatten i ett moln och fördelar det på fler droppar blir storleken på dropparna mindre. Moln med fler mindre droppar reflekterar tillbaka mer solljus till rymden än moln med färre större droppar. Således gör våra utsläpp av partiklar att molnen kyler av klimatet. Hur mycket aerosolpartiklar kyler av klimatet, genom sin påverkan på moln, är den största osäkerheten i de nuvarande uppskattningarna av framtidens klimat.

Molnens droppstorlekar kan även påverka molnens nederbörd. Det är stor skillnad i storlek på regndroppar och molndroppar, det krävs ca 1 miljon molndroppar för att bilda en regndroppe. I moln med små droppar tar det längre tid innan molndropparna växer sig så pass stora att de kan bilda regndroppar. Då moln rör sig över himlen finns det en risk att fördröjd nederbörd, från moln med små droppar, till slut faller på en annan plats. Den fördröjda nederbörden kan även leda till att molnen växer sig högre och att nederbörden blir kraftigare när den väl faller. Målet med avhandlingen är att bidra till att minska osäkerheterna kring hur mycket aerosolpartiklar påverkar moln och nederbörd.

I avhandlingen har vi kombinerat markbaserade mätningar av aerosolpartiklar med satellit- och väderradarmätningar av moln. Med hjälp av de markbaserade mätningarna har vi kunnat uppskatta hur höga partikelkoncentrationerna i luften är. Från satellitmätningarna får vi fram hur mycket ljus molnen reflekterar och hur stora dropparna i molnen är. Med väderradar kan vi se hur mycket nederbörd som faller från molnen. Vi har även undersökt hur bra satellitmätningarna av moln är genom att jämföra dem med liknande mätningar från marken.

Vi har studerat tre olika typer av moln i avhandlingen: Låga moln, konvektiva moln och cirrus moln.

Med låga moln menas moln vars toppar inte når över 1500 m. I avhandlingen undersöks låga moln både över havet norr om Skandinavien och över Finland och Sverige. När vi undersökte hur deras egenskaper påverkas av koncentrationen av aerosolpartiklar i luften såg vi att ju fler aerosolpartiklar det fanns desto mindre var dropparna i molnen. Detta gällde både över hav och land. Molnen över hav reflekterade även mer solljus när partikelkoncentrationerna var höga. Över land däremot förändrades inte mängden solljus som molnen reflekterar när mängden aerosolpartiklar i luften förändrades. Det kan ha sin förklaring i att dynamiken i molnen också förändras när dropparna blir mindre. I molnen över land kunde vi även studera hur nederbörden förändrades när aerosolkoncentrationen varierade. Det visade sig att nederbörden var något svagare när antalet aerosolpartiklar i luften var högre.

Konvektiva moln bildas ofta när marken värms upp och värmer luften ovanför. Den varma luften stiger, kyls av och bildar blomkålsliknande moln som kan bli väldigt höga. Vi undersökte konvektiva moln över ett område runt en partikelmätstation i Finland och en i Sverige. Det visade sig att även de konvektiva molnen hade mindre droppar när mängden aerosolpartiklar i luften var hög. Dessutom, precis som för de låga molnen, visade det sig att nederbörden försvagades något när fler partiklar fanns i luften. Detta gällde främst molnen som växt sig lite högre.

Cirrusmoln är tunna slöjmoln som utgörs av iskristaller och som brukar ligga på 8-12 km höjd över marken. I undersökningen av dessa använde vi partikelmätningar från ett passagerarflygplan som man installerat mätinstrument i. Svavelpartiklarna i övre delen av atmosfären kommer inte från mänskliga utsläpp utan från explosiva vulkaner som slungar upp partiklarna högt upp i atmosfären. När vi undersökte cirrusmolnen med hjälp av satelliter visade det sig att molnen reflekterade mindre solljus de åren då mängderna svavelpartiklar på hög höjd i atmosfären var höga. Detta är i motsats till vad som förväntas för vattenmoln, vilket kan förklaras med att processen för hur ismoln bildas är mer komplicerad än bildandet av vattenmoln. Höga moln värmer klimatet mer än de kyler det, så när dessa moln tunnas ut bidrar det till att kyla klimatet.

Resultaten från avhandlingen bekräftar att molndropparna blir mindre och nederbörden försvagas när koncentrationerna av aerosolpartiklar är höga i lägre delen av atmosfären. En enhetlig slutsats angående hur mängden solljus som reflekteras av molnen påverkas av aerosolpartiklar går dock inte att dra eftersom resultaten pekar åt olika håll.

1 Research Aim

The globally averaged surface temperature of the Earth has shown a positive trend during the 20th century. It is extremely likely that the dominant cause of this temperature increase is human emissions of greenhouse gases (IPCC, 2013). The majority of the greenhouse gases are emitted from combustion processes that also result in emissions of aerosol particles. Changes in the aerosol loading of the atmosphere affect clouds because every cloud droplet and crystal start growing from an aerosol particle. The extent to which changes in aerosol loading affect clouds is currently the climate driver associated with the largest uncertainty (IPCC, 2013).

The overall aim of the research presented in this thesis is to contribute to a reduction in the uncertainties associated with aerosol-cloud-precipitation interactions by investigating how microphysical and optical properties of clouds change with aerosol number concentrations. This was accomplished by using satellite data of clouds in combination with in-situ measurements of aerosols. Meteorological parameters from reanalysis data were also used to determine how meteorological conditions influence aerosol-cloud interactions. In addition, precipitation measurements were used to investigate whether aerosols affect precipitation. The more specific goals of the thesis were:

- To validate satellite retrievals of clouds against ground-based remote sensing.
- To come up with and develop new research ideas combining a multitude of long-term open access datasets to investigate aerosol-cloud interactions.
- To develop evaluation procedures and create computer software algorithms to process satellite data in order to obtain vertical profiles of cloud properties.
- To investigate how the cloud droplet effective radius and cloud optical thickness are affected by aerosol number concentration in low-level and convective clouds.
- To examine the degree to which aerosol number concentration affect precipitation in low-level and convective clouds.
- To investigate if volcanic sulphur particles injected into the stratosphere affect the reflectance of cirrus clouds.

2 Introduction

The Earth's atmosphere is an ever changing system that helps keep the planet inhabitable by absorbing and reemitting longwave radiation emitted by the Earth. The atmosphere contains gases, aerosol particles and collections of liquid and solid hygrometers that make up clouds. Since the mid-18th century, humans have been changing the composition of the atmosphere on a large scale by emitting greenhouse gases and aerosol particles through combustion processes. The human-made increase in atmospheric greenhouse gases are thought to be the main cause of the increase in the Earth's average surface temperature observed during the last century (IPCC, 2013). The emissions of aerosol particles have, however, masked some of the heating caused by the greenhouse gases, as these particles tend to cool the climate.

2.1 Aerosol particles

An aerosol is defined as solid or liquid particles suspended in a mixture of gases. The particles are either emitted at the source as particles (primary production) or emitted as vapours. The vapours can later either condense on existing particles or nucleate and form new particles (secondary production). The size of the aerosol particles range from roughly 3 nm to 100 µm. The lower size limit is set to where a stable cluster of molecules forms and the upper limit to the size where particles can no longer stay airborne. The residence time for aerosols in the troposphere is in the order of a week (Balkanski et al., 1993). Aerosol number concentrations depend very much on location and altitude. Over remote oceans concentrations are typically 100-300 cm^{-3}, while in rural continental areas the concentrations can vary between 1000 and 10000 cm^{-3} (Seinfeld and Pandis, 2006). In cities, the concentrations can be even higher than this.

The sources of aerosol particles can either be anthropogenic or natural. Anthropogenic combustion processes produce both primary particles, such as soot, and secondary particulate mass from emissions of, for example, volatile organic compounds (VOC), SO_2, and NO_x (Seinfeld and Pandis, 2006). Natural sources usually produce large primary particles, for example dust from desserts or sea spray aerosols from the oceans. However, some natural sources such as forests produce secondary aerosols of smaller sizes. Other natural sources of aerosols that can produce very high concentrations of aerosol particles are forest fires and volcanoes. Explosive volcanoes can inject gases and particles into the stratosphere where their residence time can be up to a few years. An increase in stratospheric aerosol loading between 2005 and 2009 can be linked to volcanic eruptions (Vernier et al., 2011; Friberg et al., 2014).

The different mechanisms through which aerosol particles affect the climate have been divided into the direct aerosol effect and the indirect aerosol effect. The direct aerosol effect refers to the reflection and absorption of both incoming shortwave and outgoing longwave radiation by aerosols in the atmosphere. The reflection of shortwave radiation is larger than the absorption of longwave radiation and the net aerosol effect on climate is hence negative. The increase in aerosol loading since 1750, due to human activities, is estimated to have produced a radiative forcing (RF) of approximately -0.27 (-0.73 - 0.27) Wm^{-2} according to the Fifth Assessment Report by the IPCC (2013), see Figure 1. The large uncertainty is partly caused by the short atmospheric lifetime of the aerosol particles. The short lifetime induces large spatial variation in the aerosol loading, which is much higher closer to the sources than in remote regions with few sources. The large spatial variation means that the aerosol effect on climate varies significantly between different regions of the globe (Jacob, 1999; Boucher et al., 2013). Moreover, particles of different composition have different radiative properties and hence affect the climate differently. A further complicating factor is that the size and properties of the aerosol particles change in the atmosphere (IPCC, 2013).

The net first direct aerosol effect is negative but there are also particles that warm the atmosphere. Soot particles absorb shortwave radiation and therefore heat the atmosphere, which can stabilise it and inhibit convection (Koren et al., 2004). Aerosol particles can thus prevent cloud formation in certain conditions and thereby heat the planet.

What happens to the emissions of aerosol particles in the future will have a large effect on the future climate. Some natural sources of aerosol particles can become stronger in a warmer climate. Regions that currently are too cold to host forests could in the future become covered with these. Larger forest regions and longer growing seasons can increase the amount of aerosol particles produced from biogenic VOC, which affect the climate through both the direct and indirect aerosol effect (Paasonen et al., 2013). However, the development of the anthropogenic aerosol emissions will probably affect the climate more than changes in natural aerosol sources. Because aerosol particles cool the climate, one might think that it is a good idea to keep emitting them to counteract global warming. However, we also have to keep in mind that anthropogenic aerosol particles have a highly negative impact on human health. It is estimated that 447 000 premature deaths in the EU are caused by aerosol particles each year (EEA, 2014).

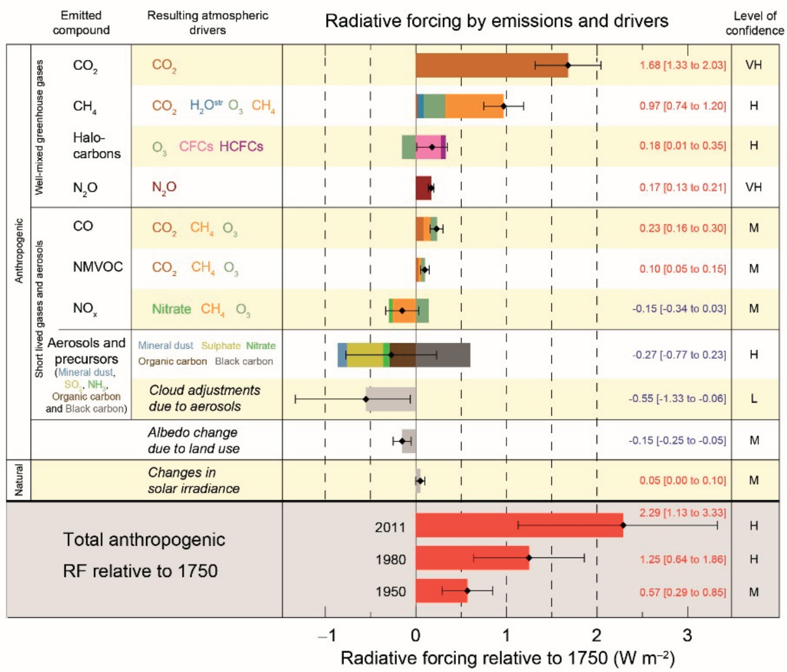

Figure 1

Radiative forcing estimates in 2011 relative to 1750 and aggregated uncertainties for the main drivers of climate change. Values are global average radiative forcing (RF14), partitioned according to the emitted compounds or processes that result in a combination of drivers. The best estimates of the net radiative forcing are shown as black diamonds with corresponding uncertainty intervals; the numerical values are provided on the right of the figure, together with the confidence level in the net forcing (VH – very high, H – high, M – medium, L – low, VL – very low). Albedo forcing due to black carbon on snow and ice is included in the black carbon aerosol bar. Small forcings due to contrails (0.05 W m^{-2}, including contrail induced cirrus), and HFCs, PFCs and SF6 (total 0.03 W m^{-2}) are not shown. Concentration-based RFs for gases can be obtained by summing the like-coloured bars. Volcanic forcing is not included as its episodic nature makes is difficult to compare to other forcing mechanisms. Total anthropogenic radiative forcing is provided for three different years relative to 1750 (IPCC, 2013).

2.2 Clouds and precipitation

Clouds are an important component in the Earth's climate system and affect the climate both by reflecting incoming shortwave radiation and absorbing outgoing longwave radiation. Clouds form when air cools and the relative humidity (RH) exceeds 100%. The cooling can occur when air rises to levels with lower pressure and expands (called adiabatic cooling). Clouds also form when two air masses of different temperatures are mixed, or due to radiative cooling of the air. These processes can occur at different altitudes in the atmosphere, and clouds at different atmospheric levels have different effects on the climate.

Cirrus clouds are thin clouds at high altitudes that consist entirely of ice. Because they are thin, they reflect little shortwave radiation but are nevertheless effective at trapping longwave radiation emitted from the planet. Cirrus clouds have a much lower temperature than the Earth's surface and warm the planet more than low-level clouds, which have a temperature more similar to the surface (Ackerman and Knox, 2003). In addition, the majority of the low-level clouds are thick and hence strongly reflect incoming shortwave radiation. Thus, if clouds cool or warm the planet depends on their thickness and altitude. The net effect of clouds on the climate is cooling because the reflected amount of incoming shortwave radiation is larger than the trapped amount of outgoing longwave radiation (Ackerman and Knox, 2003). There is, however, a poor understanding of how the predicted future warming of the atmosphere will affect clouds and whether clouds will reduce or enhance the rising temperatures (Boucher et al., 2013).

Clouds can be classified based on their appearance. Layered clouds, also called stratus clouds, have greater horizontal than vertical extent and often form in stable air. Convective clouds (cumulus), on the other hand, have greater vertical than horizontal extent and usually form in unstable air (Ackerman and Knox, 2003). The latter clouds often form when the ground is heated and warm air bubbles from the ground rise, cool and reach supersaturation.

Cloud droplets never form from pure water vapour in the atmosphere; the droplets need seeds to be able to form (Ahrens, 2007). Aerosol particles provide these seeds, which are called cloud condensation nuclei (CCN) for cloud droplets. Cloud ice crystals form either by the freezing of supercooled solution droplets below -38 °C, called homogeneous freezing, or the freezing is initiated by insoluble aerosol particles called ice nuclei (IN) through heterogeneous freezing. Heterogeneous freezing occurs either by the deposition of water vapour onto an IN or by the freezing of a supercooled liquid droplet caused by an IN (Pruppacher and Klett, 1997). CCN and IN concentrations in the atmosphere hence have the ability to affect cloud properties. The number of IN in the atmosphere is, however, 5 or 6 orders of magnitude lower than the CCN (Levin and Cotton, 2009). CCN can consist of various compounds, but salt containing particles can generally become CCN at lower supersaturations than particles containing organic compounds. To become IN, particles must have more specific characteristics (e.g. crystalline structure) and therefore the IN concentrations are much lower than the CCN concentrations (Pruppacher and Klett, 1997). Dust and metallic particles (Cziczo et al., 2013) have been found to work well as IN, but also soot and primary biological aerosol particles can act as IN (Hoose and Mohler, 2012).

After a cloud droplet has formed, it grows from condensation of water vapour onto the CCN. However, condensational growth is not fast enough to form rain droplets. An average rain droplet contains 1 million averaged-sized cloud droplets and it would take several days to grow a cloud droplet into a rain droplet from condensation alone (Ahrens, 2007). Thus, other processes are required to produce precipitation. In clouds with only liquid water, collision-coalescence processes are responsible for the droplet growth. Large droplets may form on large CCN or by the random collision of small droplets. If the droplets become large enough, they will start to fall and collide with smaller droplets, which causes them to grow even larger. The droplets can then become large enough to produce precipitation (Barry and Chorley, 2003).

Warm rain from clouds consisting entirely of water exists mainly in the tropics and subtropics, while at mid- and high latitudes, mixed-phase processes are almost always responsible for the formation of precipitation (Field and Heymsfield, 2015). In mixed-phase clouds, the interactions between the liquid and solid phase cause the ice crystals to grow. The saturation vapour pressure is higher over water surfaces than over ice surfaces, which causes the ice crystals to grow at the expense of the water droplets. This is called the Wegener-Bergeron-Findeisen process. When the ice crystals grow large enough they can start to fall and collide with droplets or other ice crystals and grow even bigger. Sometimes, when large ice crystals collide, they break into smaller fragments and create new, small ice crystals (Pruppacher and Klett, 1997).

For clouds to produce precipitation, they need to be thick enough to allow a sufficiently long fall time for the large droplets and/or ice crystals. If the droplets/crystals have time to collide with enough other droplets they can grow into precipitation-sized droplets/crystals (Levin and Cotton, 2009). Updrafts in the clouds can also prolong the sedimentation time and provide favourable conditions for clouds to produce precipitation. Even if precipitation-sized particles form in the clouds, they often evaporate when falling through a dry atmosphere below the clouds and hence no precipitation reaches the ground. In a similar fashion, ice crystals often melt while falling through the clouds or the atmosphere below and reach the surface as rain.

2.3 Aerosol-cloud-precipitation interactions

As mentioned in section 2.1, how anthropogenic emissions of aerosol particles affect climate by changing cloud properties is called the indirect aerosol effect. Human activities, such as combustion, have increased the amount of aerosol particles in the atmosphere since the Industrial Revolution. Clouds that form in air with more particles present will have more droplets than those that form in clean air, and more droplets imply smaller droplets, if the amount of liquid water in the clouds stays the same. It has long been known that clouds with many small droplets reflect more sunlight than clouds with few large droplets if they have the same liquid water content (Twomey, 1974). A change to smaller droplets in the clouds can also affect how rapidly the clouds produce precipitation and may suppress light rain (Albrecht, 1989). Suppressed drizzle can increase the cloud lifetime and thereby cause further cooling of the climate.

In Figure 1, it can be seen that the indirect aerosol effect is associated with a RF of approximately -0.55 (-1.33 to -0.06) Wm^{-2} (IPCC, 2013). The coarse resolution of the global climate models and the up-scaling of the interactions between aerosols and clouds are factors that contribute to the large uncertainty in the RF. Moreover, different types of clouds are more or less sensitive to aerosol perturbations. Another factor contributing to the large error bars are the difficulties in estimating the aerosol loading of the pre-industrial era (Carslaw et al., 2013). Investigations have shown that clouds are more sensitive to the size and number of aerosol particles present in the atmosphere than to their chemical composition (Andreae and Rosenfeld, 2008).

Marine low-level clouds are considered to be very sensitive to small increases in CCN concentrations because they normally form in very clean environments and the aerosol effect on cloud droplet number concentration (N_d) have been found to saturate at high particle concentrations (Verheggen et al., 2007). Low-level clouds cover approximately 23% of the ocean surface (Warren et al., 1988). Because of the large contrast between the dark ocean background and the clouds, a small change in cloud reflectivity or lifetime of marine low-level clouds can significantly affect the Earth's radiation budget. Furthermore, in low-level clouds, drizzle can be suppressed by increasing aerosol loading since it takes small cloud droplets longer to grow into drizzle-sized droplets.

However, investigations of aerosol effects on low-level clouds have come up with varying results. Most studies find that the droplets are smaller in clouds formed in more polluted conditions (e.g.Twohy et al., 2005; Costantino and Breon, 2013). But the results regarding whether smaller droplets actually result in clouds that reflect more sunlight show more inconsistency. Some studies find that the low-level clouds formed in more polluted environments have a higher cloud optical thickness (COT) (a measure of how much light that is reflected by the clouds), than those formed in cleaner conditions (Chameides et al., 2002; Guo et al., 2007). Yet other studies have found no changes in COT with aerosol number concentrations (Twohy et al., 2005; Costantino and Breon, 2013). One cause for the diverging results may be that air masses with different aerosol loading often also have different origins. Continental air masses are often associated with both higher aerosol particle concentrations and drier conditions, which results in clouds containing less water (Brenguier et al., 2003). Another reason for the varying results regarding aerosol effects on COT is that the decrease in droplet size in polluted conditions can affect the dynamics of the clouds. Smaller droplets can lead to enhanced mixing of dry air into the clouds. This lowers the amount of water in the clouds and hence the COT (Rosenfeld et al., 2014). The amount of sunlight reflected by the clouds is more sensitive to changes in liquid water path (LWP) than changes in N_d concentration (Boers and Mitchell, 1994; Boucher et al., 2013).

Convective clouds are also sensitive to changes in aerosol loading and are often fed aerosol-rich boundary-layer air through their base. Increased aerosol loadings are thus associated with decreased droplet sizes at the base of the convective clouds. In clouds with warm cloud bases, the smaller droplet sizes are thought to result in a suppression of warm precipitation, which leads to more cloud water being transported to freezing altitudes. This results in a shift in the latent heat release in the clouds and leads to an invigoration of the clouds. The initiation of the precipitation is delayed but the precipitation that later falls may be more intense than it would have been from a cloud formed in a pristine environment (Rosenfeld et al., 2008a). Nonetheless, the sensitivity of convective clouds to aerosol perturbations have been found to depend on a range of meteorological conditions such as vertical wind shear (Fan et al., 2009), atmospheric instability (Lee et al., 2010) and RH close to the cloud base (Khain et al., 2008).

Cirrus clouds are mainly sensitive to the amount and distribution of IN in the atmosphere (Kärcher and Spichtinger, 2009). How the cirrus clouds respond to changes in IN concentrations depends on the freezing mechanism that is responsible for the cirrus ice crystals. If homogeneous nucleation dominates the cirrus formation, adding a few IN can seed the clouds and shift the nucleation mechanism from homogeneous to heterogeneous. This results in clouds with fewer larger ice crystals and these clouds have a lower COT. Since cirrus clouds absorb more longwave radiation than they reflect shortwave radiation, a decrease in the COT of cirrus clouds would cool the climate. If, on the other hand, heterogeneous nucleation is the dominating freezing mechanism, an addition of IN would lead to more, smaller ice crystals and optically thicker clouds. This would result in a warming of the climate.

Clouds also affect aerosols. Wet scavenging through precipitation is one of the most effective ways of removing aerosol particles from the atmosphere. However, most clouds do not produce precipitation and the majority of the cloud droplets do not become rain droplets. When the cloud droplets instead evaporate, the aerosol particle distribution looks different than it did before the cloud processing. Chemical processes occurring in the droplets, as well as coalescence between droplets or droplets and interstitial aerosol particles (particles that failed to become cloud droplets) (Seinfeld and Pandis, 2006) will most often reduce the number of particles and make them larger.

3 Method

3.1 MODIS cloud retrievals

Data from the passive radiometer MODIS (Moderate Resolution Imaging Spectroradiometer) have been used in all five papers included in this thesis. The MODIS instruments are placed on the Terra satellite, which was launched in December 1999, and the Aqua satellite, launched in May 2002 (Platnick et al., 2003). Both satellites are polar orbiting at an altitude of 705 km. Terra has a descending orbit with an equatorial passing time of 10:30 local solar time while Aqua has an ascending orbit and passes the equator at 13:30.

The MODIS instrument is a whiskbroom scanning radiometer with a swath width of 2330 km. The swath is divided into segments called granules that are 1354 x 2330 km large. MODIS has 36 spectral bands (also called channels) that cover the wavelengths from 0.415 to 14.235 μm (King et al., 2003; Platnick et al., 2003) and the bands have spatial resolutions of 250 m (2 bands), 500 m (5 bands) and 1000 m (29 bands) at nadir. Each MODIS instrument manages a global coverage every two days (King et al., 2003) but areas closer to the poles are covered two times a day or more often by each satellite.

The MODIS team provides a range of different atmospheric products that are derived from the data collected at the different wavelength bands. The calibrated data from the wavelength bands are called level 1B products, while the atmospheric products calculated from these are called level 2 products. There are also level 3 products, which have global coverage with 1 by 1° latitude-longitude resolution and are available with different temporal resolution (King et al., 2003). The level 1B, 2 and 3 products have been produced in different collections with updates and improvements being made with each new collection. In Paper I, collection 6 and 5.1 were compared. Collection 5 was utilised in Paper II, and in Papers III and IV, collection 5 was used for Terra and collection 5.1 for Aqua. In Paper V, collection 5.1 was employed.

3.1.1 Level 1 products

Level 1B products were utilised in the study of convective clouds (Paper IV) to calculate the cloud top temperature (CTT) at 1 km spatial resolution rather than using the CTT provided in the level 2 data from collections 5 and 5.1 (5 km resolution). Data from two near-infrared bands centred at 11 and 12 μm were used in the CTT calculations.

3.1.2 Level 2 cloud products

The two main level 2 cloud parameters used in this thesis research to determine the effects aerosol particles have on clouds were the cloud droplet effective radius (r_e) and the COT. r_e is a measure of the cloud droplet sizes and is defined as the third moment of the droplet distribution divided by the second moment.

$$r_e = \int_0^\infty r^3 n(r)dr / \int_0^\infty r^2 n(r)dr \qquad (1)$$

where n(r) is the frequency function of the cloud droplet sizes. COT is a measure of how much of the incoming radiation that penetrates a cloud.

$$I = I_0 e^{(-COT)} \qquad (2)$$

where I_0 is the incoming intensity of the light and I is the outgoing intensity.

These two parameters are simultaneously retrieved using the reflectance from one non-absorbing band (visible) and one water-absorbing band (near-infrared or infrared) (Platnick et al., 2003). Three different visible bands are used over different underlying surfaces: one for ocean, one for land, and one for ice/snow surfaces. Moreover, three different water absorbing bands (1.6 μm, 2.1 μm and 3.7 μm) are used to produce three different r_e products. The COT and r_e are obtained from the reflection obtained by MODIS in the two chosen wavelength bands by using a look-up table approach. The look-up table is created using a radiative transfer model to calculate the amount of reflection in different wavelength bands produced by clouds of different COT and r_e. The sensor and solar angles as well as the altitudes of the clouds and several other parameters that can affect the reflection from the clouds are also considered in the calculations of these look-up tables (King et al., 1997; Platnick et al., 2003). The retrieved reflectance from MODIS can then be matched against the look-up tables to determine the COT and r_e. An example of a look-up table of liquid clouds is presented in Figure 2.

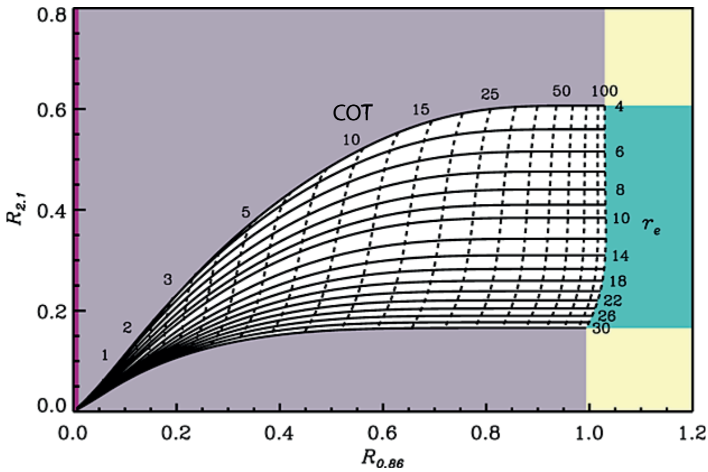

Figure 2
An example of the retrieval solution space for a liquid-phase cloud over an ocean surface, assuming the solar zenith angle is 20°, the sensor zenith angle is 20°, and the relative azimuth angle is 0°. The reflectance in the 0.66 µm and 2.1 µm channels are shown on the axis (Cho et al., 2015).

Retrieving cloud parameters with the look-up table approach is associated with certain problems. The solutions for r_e become uncertain at low COT as can be seen in Figure 2. In Papers III and IV, only pixels with COT greater than 5 and 7 were used. Another problem with look-up tables is that the algorithm assumes that the clouds are plane parallel which is certainly not true for all clouds, especially not convective clouds. The plane-parallel problem can increase at higher latitudes since the solar zenith angles are low there (Grosvenor and Wood, 2014). Another problem with the look-up table approach is that it assumes that clouds are either liquid or ice clouds. Each pixel containing clouds is defined as liquid or ice and separate look-up tables exist for the different phases (King et al., 1997). Most clouds in the atmosphere with temperatures between -6 °C and -38 °C contain both supercooled water and ice. Hence, assuming that all clouds are either liquid or ice is somewhat unrealistic.

Different MODIS r_e products retrieved from different wavelengths were utilised in the research on which this thesis is based. In Paper II, the standard 2.1 µm r_e was used, while in Papers III and IV, the r_e at 3.7 µm was analysed. In recent years, research has shown that the r_e at 3.7 µm is less sensitive to problems with the plane-parallel cloud assumption (Zhang et al., 2012), which is why it was used in Papers III and IV.

Another cloud product that has been utilised in this study is the LWP which is a measure of how much condensate is present in the clouds. The LWP is not retrieved separately but calculated from the COT and r_e through the following formula:

$$LWP = 4\, COT\, r_e / 3\, Q(r_e) \qquad (3)$$

where $Q(r_e)$ is the extinction efficiency in the visible band used in the retrievals of COT and r_e (King et al., 2006). The standard r_e at 2.1 µm is used in the derivations of LWP.

The CTT and cloud top height (CTH) measurements were used in the research to determine the level in the atmosphere where the clouds were placed. Another cloud property, cloud top pressure (CTP), is closely connected to the other two but has not been directly used in this research. Two different retrieval algorithms are used by the MODIS CTP retrievals. The first is called the CO_2 slicing technique and uses ratios between radiance from channels in the 15 µm region to determine the CTP. However, this technique does not work for low-level liquid clouds; for these a technique called the infrared window approach (IRW) is used. The IRW use the brightness temperature (BT) in the 11 µm channel to retrieve the CTT. Both methods are then combined with gridded meteorological data from the Global Data Assimilation System (GDAS) to convert the pressure to temperature and vice versa (Menzel et al., 2008). The CTH is also obtained by combining CTP or CTT with meteorological data but is only available in collection 6, the latest collection of cloud products (Baum et al., 2012). In collection 6, the resolution of the cloud top properties was also increased to 1 km spatial resolution at nadir. The CTT, CTP and CTH are produced from both daytime and nighttime data, but the optical properties (r_e, COT and LWP) require daylight and are thus only produced during the day.

3.1.3 Level 3 cirrus reflectance

The MODIS level 3 products are provided at daily, eight-day and monthly temporal resolutions. In this thesis research, only the monthly resolution product was used.

MODIS band number 26 centred at 1.38 µm was specifically chosen for the study of cirrus clouds. The incoming solar radiation at this wavelength is either scattered by the cirrus clouds or totally absorbed by the atmosphere below. There is, however, some attenuation of the reflection due to water vapour located above the cirrus clouds (King et al., 2003). To account for this, each granule is divided into 16 subgranules and scatter plots of the reflectance in the band at 1.38 µm and the band at 0.65 µm are created using the pixels in each subgranule. A scaling factor is calculated from each scatterplot and the scaling factors are interpolated to avoid a chessboard effect (Meyer and Platnick, 2010). The scaling factors are then used when calculating the cirrus reflectance (CR) for each pixel.

When the atmosphere below the cirrus clouds is dry, some of the reflectance from lower clouds or the surface can contaminate the CR. To remove such data, the atmospheric water vapour product (or column precipitable water) from MODIS was used. The unit of the products is cm, which is the height that the column water vapour would reach if it was condensed to liquid water.

3.1.4 Cloud profiles

A method developed by Rosenfeld and Lensky (1998) was implemented in Paper IV to obtain vertical profiles of clouds from satellite scenes. Convective clouds in a satellite scene extend to varying heights and thus their cloud tops will have different temperatures (Figure 3a). It is assumed that the different cloud tops represent clouds at different development stages, which has been confirmed by both satellite investigations (Lensky and Rosenfeld, 2006) and aircraft measurements (Freud et al., 2008a). The CTT is plotted on the y-axis and the r_e or the COT on the x-axis to obtain vertical profiles of these cloud properties, see Figures 3b and 4. The y-axis in Figure 4 is reversed so that the coldest cloud pixels end up in the uppermost part of the figure. This approach has mainly been used to obtain profiles of the r_e (Rosenfeld and Lensky, 1998; Lin et al., 2006; Freud et al., 2008b) but a few studies have also used it to get the vertical profiles of the COT, for example, Koren et al. (2005).

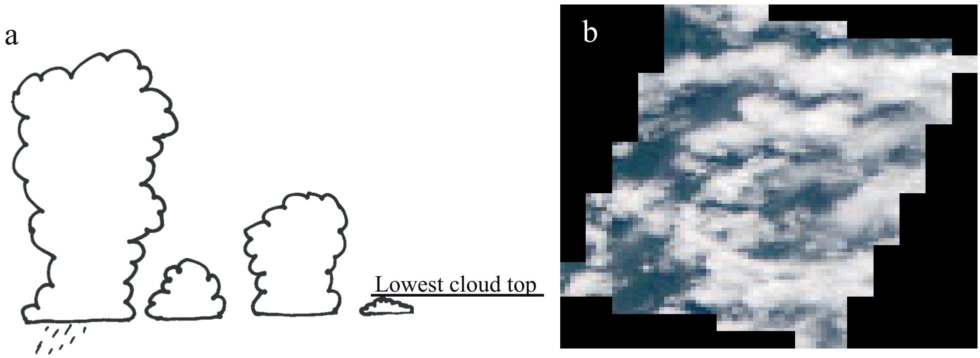

Figure 3
a) Clouds with different CTT that represent clouds at different development stages. The lowest cloud top will become the base of the profile created from this image. b) Satellite image over central Finland displaying convective clouds. Whiter clouds mean thicker clouds.

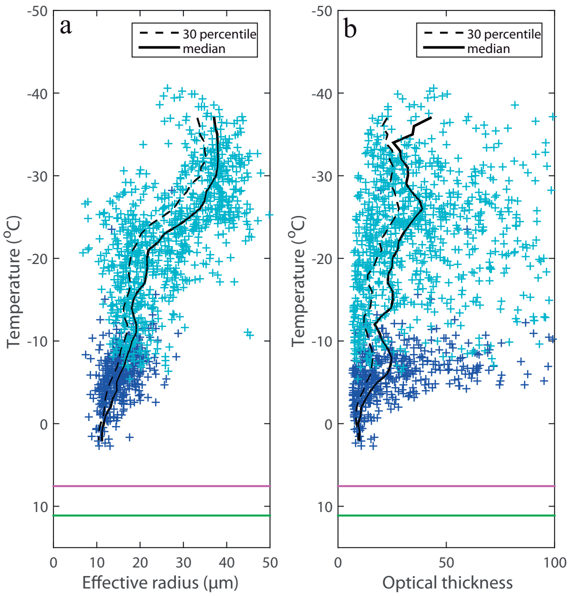

Figure 4
Vertical profile created for the satellite scene in Figure 3b. a) shows the vertical profile of the r_e, while b) shows the vertical profile of the COT. Navy blue crosses represent water pixels while cyan crosses represent ice pixels. The green horizontal line marks the surface temperature while the magenta horizontal line marks the temperature at the lifting condensation level.

The vertical profiles of the r_e can be used to find evolution stages (zones) of convective clouds which are defined in Rosenfeld and Lensky (1998). The zones will be briefly described here, and Figure 5 shows an r_e profile containing all the intervals. At the bottom of the cloud is the diffusional droplet growth zone, which is characterised by a small increase of r_e with height because the droplets grow slowly here. Above this zone is the coalescence growth zone where r_e increases greatly with altitude at the level of the cloud where the temperatures are greater than zero. Next is the rainout zone, where r_e is constant with height at 20 to 25 μm. In this zone, the droplets become big enough to overcome the updrafts and start falling, which balances the coalescence and causes a constant r_e. The fourth zone is the mixed-phase zone where the r_e grows rapidly with height again, but this time at freezing temperatures due to mixed-phase precipitation processes. The last zone is the glaciated zone where r_e is once again stable, but at larger sizes than in the rainout zone. The size of the ice crystals is determined by the strength of the upward winds. All five zones described here are not always present in convective clouds.

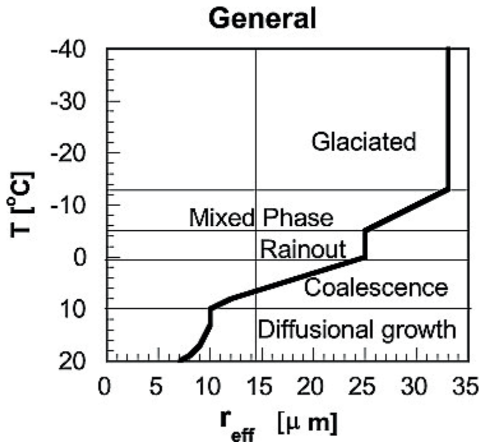

Figure 5
Classification scheme of convective clouds into microphysical zones according to the shape of the CTT-r_e relations (Rosenfeld et al., 2008b). Republished with permission from the American Geophysical Union.

The vertical r_e profile in Figure 4a displays a long diffusional droplet growth zone but since there is no rapid increase in r_e at temperatures above freezing, no droplet growth or rainout zone is present. At -20 °C, the r_e starts growing rapidly at the mixed-phase zone and the glaciation zone is visible at altitudes with temperatures below -30 °C. The coalescence and rainout zones were hardly ever present in the cloud profiles in Paper IV. Clouds at the latitudes of the study seldom form at temperatures warm enough to allow for warm droplet coalescence growth and a rainout zone. It is nevertheless quite common that the rapid growth starts at temperatures above freezing level and then continues into the mixed-phase zone without entering the rainout zone.

Not all pixels in the satellite scenes are included in the build-up of the profiles. Firstly, the pixels have to contain clouds to be included in the analysis. Moreover, when pixels contain several layers of clouds, the retrieval of the CTT, the microphysical and the optical properties become uncertain. That is why these cloud pixels are not included in the analysis. Cirrus clouds are often produced by large convective towers, but can also be present in the satellite scenes due to other large scale meteorological conditions. Nevertheless, since cirrus clouds are thin but very cold they will lower the COT at the top of the profiles. This does not represent the different evolution stages of a convective clouds and pixels containing cirrus and multilayer clouds were thus excluded from the profile analysis using the MODIS multilayer cloud warning and CR products.

Selection criteria were applied to the profiles as well. The droplet sizes in convective clouds increase with height (Rosenfeld and Lensky, 1998) and hence the r_e in the profiles needs to increase with height to be included in the analysis. As clouds grow taller they will increase in thickness and their COT will also increase; hence profiles need to have an increasing COT to be included in the dataset. In the profiles that do not fulfil these two criteria, other types of clouds that disturb the profile retrievals are most likely present in the satellite scene. The lifting condensation level is the height where convective clouds are expected to start forming according to the temperature and humidity at the surface. Profiles with bases considerably higher up in the atmosphere than the lifting condensation level were excluded from the analysis.

3.2 VIIRS cloud retrievals

The Visible Infrared Imaging Radiometer Suite (VIIRS) sensor is placed on the Suomi-NPP satellite which was launched in October 2011. The satellite is polar orbiting in a sun-synchronous ascending orbit that crosses the equator at 13:30 local solar time. VIIRS has 16 channels with a 750 m spatial resolution and 6 channels with a 350 m spatial resolution at nadir (Cao et al., 2013). VIIRS has been designed to be similar to MODIS and will continue similar Earth system monitoring after the MODIS satellites are taken out of use.

CTH, a VIIRS level 2 cloud product, was used in Paper I of this thesis. The Level 2 data were produced using software from the Polar Platform System (PPS) and the level 1 data were produced at the Swedish Meteorological and Hydrological Institute (SMHI) from locally received VIIRS data. Different algorithms were applied to opaque clouds and semi-transparent (or suspected semi-transparent) clouds. For opaque clouds, the 11 μm BT is used together with gridded meteorological data from the European Centre for Medium-Range Weather Forecasts (ECMWF) to retrieve the CTH. The method for the semi-transparent clouds is somewhat more complex and utilises the discrepancy between adjacent pixels to estimate the CTT. Meteorological data are then also applied to obtain the CTH.

3.3 Ground-based remote sensing

3.3.1 ARM Mobile Facility

The ARM (Atmospheric Radiation Measurement program - US Department of Energy) mobile facility 2, AMF2, was deployed in Hyytiälä, Finland, as part of the measurement campaign BAECC (Biogenic Aerosols – Effects on Clouds and Climate) from February to September 2014. AMF2 contains a comprehensive suite of ground-based in-situ instrumentation together with active and passive remote-sensing instruments to obtain numerous atmospheric properties with very high temporal and spatial resolution. In Paper I, CTH and LWP obtained from AMF2 were compared to their satellite counterparts. CTH is provided by the cloud mask created from a combination of the 35-GHz KAZR (Ka-band ARM Zenith-pointing cloud Radar) and the micropulse lidar or ceilometer. Gaps in the operation of the KAZR instrument were supplemented by the 95-GHz MWACR (Marine W-Band ARM Cloud Radar). These instruments were processed using the Cloudnet scheme (Illingworth et al., 2007) which diagnoses the atmospheric targets (such as aerosol, cloud, or precipitation) together with their phase if appropriate. CTH is then obtained directly as the highest cloud pixels diagnosed by the target classification. LWP is obtained from the Radiometrics microwave radiometer (MWR); a passive instrument measuring the microwave BT at 23.8 and 31.4 GHz. Column-integrated water vapour and liquid water amounts are obtained through the use of a radiative transfer model using the monthly regression coefficients (Liljegren, 1999).

3.3.2 Weather radar – BALTEX

Radar data from the Baltic Sea Experiment (BALTEX) were used to investigate the precipitation rate of the clouds. The aim of BALTEX was to investigate the hydrological cycle and the exchange of energy between the Earth surface and the atmosphere in and around the Baltic Sea. One part of BALTEX is the BALTEX Radar Network (Michelson et al., 2000) that incorporates weather radars in 7 countries around the Baltic Sea. The composite images produced have a 2 km spatial resolution. An image containing the radar reflectivity factor in dBZ is available every 15 minutes, which means that the maximum separation between the satellite image and its respective radar image is 5 minutes. Moreover, the accumulated precipitation over 3 hours at the same horizontal resolution is calculated from the radar reflectivity data by the BALTEX Radar Data Centre.

3.4 Ground-based in-situ measurements

3.4.1 Aerosol particles

Several measurement stations for aerosol particles have been used in the thesis research. In Paper II, data from three Finish aerosol stations were investigated: Värriö (Hari et al., 1994; Vehkamaki et al., 2004), Pallas (Lihavainen et al., 2003) and Hyytiälä (Hari and Kulmala, 2005). The data from the stations were used to estimate aerosol concentrations in air masses moving out over, or in from, the oceans north of Finland. In Papers III and IV, data from Vavihill in southern Sweden (Kristensson et al., 2008) and, once again, data from the measurement station in Hyytiälä were analysed. The satellite studies were focused over smaller regions around the two stations (Figure 6) so that no tracking of the air masses were needed. The reason that these two stations were chosen for Papers III and IV were that they both have long measurement series of aerosol particles in addition to homogeneous surroundings well-suited for satellite studies. The location of all 4 stations used in the thesis can be seen in Figure 6 and each station is further described in the respective papers.

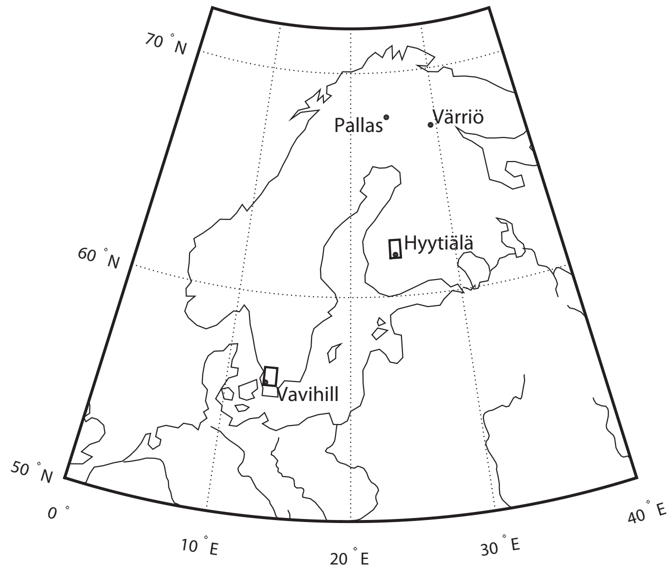

Figure 6
Map of stations used in the thesis.

Aerosol number size distributions from the differential mobility particle sizer (DMPS) were used to estimate aerosol number concentrations in Papers II, III and IV. The number of CCN was approximated by N_{80} in Paper II. This has been found to be the typical activation diameter for particles in this geographical region (Komppula et al., 2005). In the investigation of convective clouds in Paper IV, 80 nm was also found to be the activation diameter that correlated best with the cloud properties and N_{80} was used as the proxy of CCN in this paper as well. However, for low-level clouds over the same areas, N_{130} was found to be best correlated to the cloud properties (Paper III). Cloud base updraft velocities are one of the factors that control how large a fraction of the total aerosol populations becomes CCN (Rosenfeld et al., 2014). Since low-level clouds have lower cloud base updraft velocities than convective clouds, they are expected to have a higher activation diameter.

CCN counter data from Vavihill (Fors et al., 2011) and Hyytiälä were also used in Papers III and IV. However, the data are only available for 2 years and were mainly used to ensure that N_{80} and N_{130} were appropriate proxies for the amount of CCN. In the dataset in Paper IV, N_{80} correlated best with the CCN concentration at 0.4 % supersaturation for both stations. In Paper III, the ratios between CCN concentrations and N_{130} indicated that N_{130} corresponds to a supersaturation between 0.1 and 0.2 %.

3.4.2 Precipitation

Ground-based precipitation measurements have been used in Papers III and IV in order to investigate if aerosols affect precipitation. The SMHI and the Finish Meteorological Institute (FMI) both have a network of measurement stations covering Sweden and Finland, respectively. Most stations only measure precipitation on a daily basis and this was the time resolution used to include as many stations as possible within the regions studied. Hence, it is not possible to study precipitation rates using these datasets. Precipitation measurements from the Hyytiälä measurement station itself were also included. The SMHI and FMI provided the precipitation data from the stations located in the areas around the stations chosen in the satellite imagery and shown in Figure 6. In the Vavihill area there were at least 11 stations available for the entire period of time while at most, 3 stations were available for the Hyytiälä area.

3.5 HYSPLIT

The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Hess, 1997) was used in Papers II, III and IV to obtain information about air mass movements and origin. The model uses meteorological data from the GDAS model run by the US National Centers for Environmental Prediction (NCEP). In Paper II, both backward and forward trajectories were used to track the movement of the air masses whereas only backward trajectories were employed in Papers III and IV. All trajectories started 100 m above the respective stations since it was assumed that the lowest part of the atmosphere was well mixed. New trajectories were calculated every hour.

The trajectories were analysed in different manners in the papers. In Paper II, 10-day back trajectories and 4-day forward trajectories were analysed manually to determine how the air masses moved. This procedure was very time consuming and since the number of days studied were much larger in Papers III and IV, a simplified air mass background analysis was performed instead. The centre of gravity of 3-day back trajectories was calculated by taking the average latitude and longitude coordinates of the trajectories during the selected time period. The azimuth angle and the distance between the station and the centre of gravity were then calculated to determine the direction from which the air arrived at the station and how fast the air mass was moving.

3.6 ECMWF

The ECMWF, based in the United Kingdom city of Reading, is an intergovernmental organisation supported by 34 states. The Centre was established in 1975 and has been producing weather forecasts since 1979. The Centre's meteorological parameters produced by their weather models in both the forecasting and reanalysis mode have been utilised in Papers II to V.

In Paper II, monthly averages of the temperature at 1200 m for April, July and September over Barents Sea were used. From these, a function was created to screen out clouds with top temperatures that were too cold so that only low-level clouds were studied. The wind speed at 2 m altitude over the same region was used to investigate if sea salt aerosols affected the aerosol number concentration during the movement of the air masses between the station and the ocean region investigated.

In Paper III, the ECMWF profiles of the temperature and specific humidity (SH) above the Vavihill and Hyytiälä areas were used to calculate the temperature at 1500 m for each case. This was done to be able to screen out clouds at higher levels. The humidity conditions in the boundary layer were investigated using the RH and SH at 1000 hPa. The lower-tropospheric static stability (LTSS), which is a measure of the stability in the lower atmosphere, was also calculated from the ECMWF data.

In Paper IV several different ECMWF meteorological parameters were utilised to determine how these affected the clouds and precipitation. The RH and SH at 1000 hPa were used to investigate how the humidity of the air affects the clouds. The instability of the atmosphere was estimated by convective available potential energy (CAPE) and the vertical winds at 500 hPa. From the temperature and dew point measurements at 2 m, the lifting condensation levels of the clouds were also calculated.

In Paper V, ECMWF data were used to calculate the potential vorticity (PV) along the flight path of the aircraft.

3.7 CARIBIC

The CARIBIC (Civil Aircraft for Regular Investigation of the atmosphere Based on an Instrument Container) project uses a passenger aircraft carrying an instrument container to investigate aerosol particles and trace gases in the lowermost stratosphere (LMS) and upper troposphere (UT) (Brenninkmeijer et al., 1999; Brenninkmeijer et al., 2007). The project has been ongoing since 1997 with a break between May 2002 and November 2004 when the instrument container was scientifically extended and moved to a different aircraft. The flights originate in either Munich or Frankfurt and fly to destinations in Asia, Africa and North and South America. The air is sampled through inlets placed underneath the aircraft, in front of the engines. The aircraft cruising altitude of 9-12 km and the varying tropopause height enable sampling of air from both the LMS and UT.

The instrument container contains an impactor-based aerosol sampler which collects particles in the size range 0.08-2 μm (Nguyen et al., 2006). The sampler was improved during the 2002-2004 break. The collection time per sample is approximately 100 (150) minutes for the new (old) impactor, which corresponds to a distance of approximately 1500 (2200) km (Friberg et al., 2014). The aerosol samples are collected on thin polyimide films and then analysed for elemental composition at the Lund Ion Beam Analysis Facility by combining two IBA (ion beam analysis) techniques, particle induced X-ray emission (PIXE) and particle elastic scattering analysis (PESA) (Martinsson et al., 2014).

4 Results and Discussion

4.1 Satellite cloud product validation

In Paper I of this thesis, satellite remote sensing of cloud properties were compared with ground-based remote sensing. The ground-based data came from the deployment of the AMF2 in Hyytiälä, Finland between February and September 2014. The satellite instruments used were MODIS aboard the Terra and Aqua and VIIRS aboard the Suomi-NPP satellite. The VIIRS data were only compared from February to the beginning of May but the MODIS data were available for the entire duration of the ground-based campaign. A circle with a diameter of 30 km in the satellite data was compared to 1 hour of ground-based data centred at the overpass of the satellite.

The comparison of the CTH is shown in Figure 7 for daytime a) and nighttime b) conditions. Since the satellite retrievals become more uncertain when several layers of clouds are present, the results were divided according to if there were multilayered clouds present or not in the ARM data. The median differences between the satellite and ARM CTHs for the single layer dataset were generally positive by a few hundred meters. When the whole dataset was included, the differences decreased and tended to become negative by a similar magnitude. The CTH for high clouds is often underestimated by satellites and mainly high-level clouds were removed by the multilayer filter. This may be one reason for the median differences becoming more negative for the entire dataset. However, the MODIS retrievals may also underestimate the CTH when multiple layers of clouds are present. The standard deviations of the CTH differences were high and there were quite a few outliers for which the satellite retrievals underestimated the CTH by several thousand meters. Nevertheless, the median differences indicate that the satellite-derived CTHs agreed reasonably with their ground-based counterparts and most of the data points were fairly close to the 1:1 line.

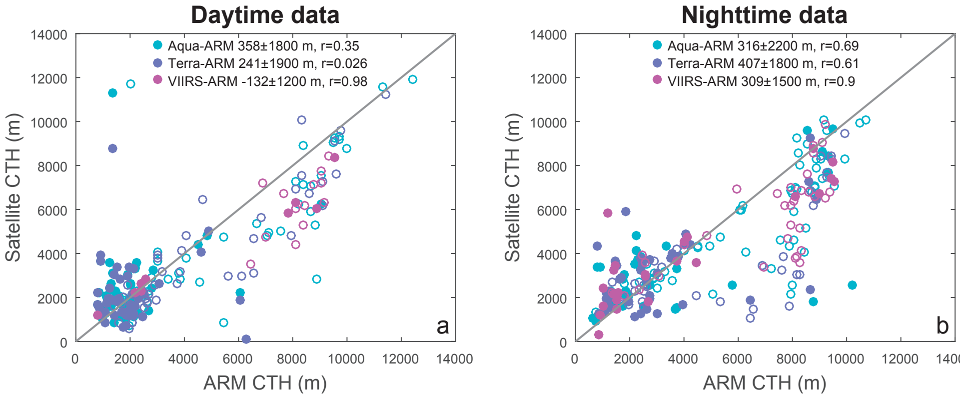

Figure 7
Comparison of CTH between ground-based measurements and satellite for a) daytime data, and b) nighttime data. The dataset is divided according to whether the ARM data contain multilayered clouds (open) or not (solid). The legend displays median differences, standard deviations and correlation coefficients between the datasets with only single layer clouds.

The second parameter compared in Paper I was the LWP and for this parameter only MODIS data were compared. Not enough VIIRS data were available for a comparison since only liquid clouds were included and the VIIRS data were only available for the first three months of the campaign when it was both dark (daylight needed for the retrieval) and cold (mainly ice clouds formed). The multilayer cloud flag (only available during the day) from MODIS was utilised to screen away multilayer clouds from the satellite scenes and all multilayer ARM data were also removed.

Figure 8 contains the results for the LWP for a) collection 5.1 and b) collection 6. The LWP was underestimated by the Terra satellite in collection 5.1 (-12.1 gm^{-2}, -14.3 %) and somewhat overestimated by Aqua (5.03 gm^{-2}, 4.56 %). For collection 6, both satellites overestimated the LWP (Aqua 12.0 %, Terra 10.4 %). There has been a small negative drift in the reflectance of the MODIS instrument aboard Terra, which has been corrected for in collection 6 (Sun et al., 2012). This may have been the cause of the negative differences between the Terra collection 5.1 and ARM data. Apart from this, there did not appear to be a very large difference in the performance of the collections. The differences between the satellites and the ground-based remote sensing increased significantly as the LWP exceeded 200 gm^{-2}. The data below 200 gm^{-2} however agreed well between the satellite and ground-based measurements which is reassuring since the MODIS LWP used in Papers II and III are mainly in this range.

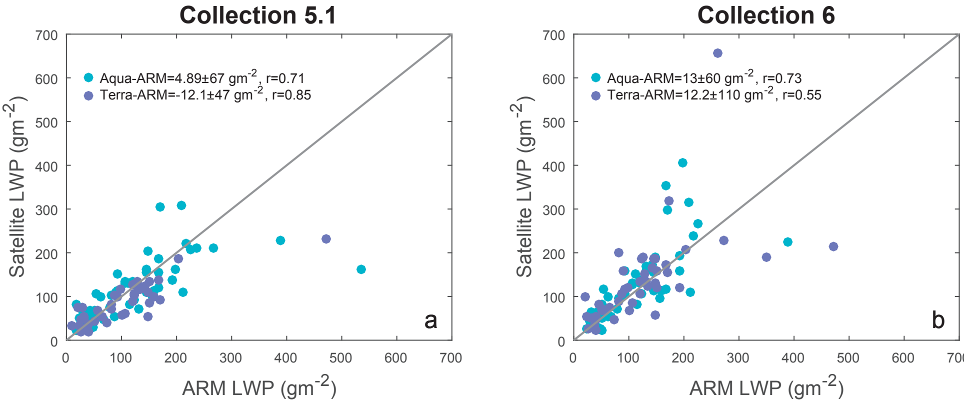

Figure 8
Comparison of LWP between satellite and ground-based measurements for a) MODIS collection 5.1 and, b) MODIS collection 6. The legend displays median differences, standard deviations and correlation coefficients between the datasets.

4.2 Low-level clouds

In Papers II and III, aerosol effects on low-level clouds were investigated. Marine low-level clouds over the Barents Sea north of Scandinavia were investigated in Paper II while continental low-level clouds over Sweden and Finland were the topic of Paper III. In Paper II, HYSPLIT was used to track air masses out over the ocean (polluted), or in from the ocean (clean) to measurement stations in northern Finland (Figure 6). This was done in order to estimate CCN concentrations over the ocean. Only the most polluted (highest 25th percentile of N_{80}) and cleanest (lowest 35th percentile of N_{80}) of the days during a 5-year period were investigated. The pollution limits, tracking and cloud conditions restricted the number of cases and in total 22 clean and 25 polluted cases were included in the study.

In Paper III, CCN concentrations were estimated from ground-based stations placed inside the areas investigated in the satellite images, and no tracking of air masses was carried out. At Vavihill, investigations of 9 years of data resulted in 122 days while at Hyytiälä, 10 years were investigated of which 261 days were included in the study.

4.2.1 Effective radius

High aerosol loading was associated with low r_e in the research presented in both Papers II and III. This agrees with the hypothesis that increased aerosol loading results in more, smaller droplets. In Paper II, all the pixels from satellite scenes recorded in clean conditions were compared to all the pixels from polluted conditions (Figure 9a). The cloud droplets formed in air with higher CCN concentrations had a significantly lower geometrical average r_e (10.7 µm) than those formed in cleaner conditions (14.4 µm). The relationship between r_e and N_{100}, a proxy for CCN, is shown in Figure 9b. In Paper II, N_{80} was used as the CCN proxy and in Paper III, N_{130} was found to best correlate with cloud properties. In order to compare the results between the two papers, N_{100} is used in Figure 9b. The r_e is seen to decrease with increasing aerosol loading for all three locations but the marine low-level clouds have higher r_e. The standard r_e product from MODIS (2.1 µm) was used in Paper II while the r_e at 3.7 µm was used in Paper III. However, using the 2.1 µm r_e for the Paper III data only slightly raised the r_e values from Vavihill and Hyytiälä. Hence, the different r_e products were not the cause of the larger r_e values over the ocean. The cause was instead thought to be the different dynamical and/or thermodynamical conditions over the respective areas. The powers of x (Figure 9b) for the different locations did, nonetheless, have similar values.

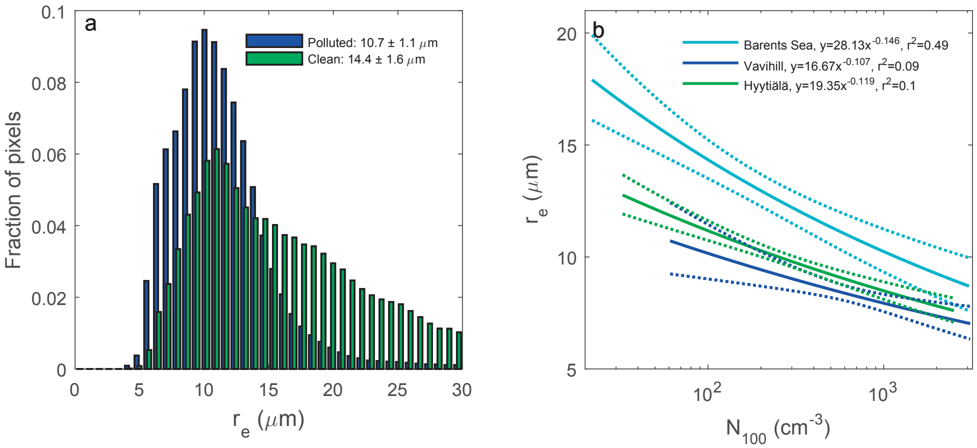

Figure 9
a) Normalised r_e distributions of the polluted and clean pixels and geometrical averages of the distributions with corresponding uncertainties, from Paper II. b) Logarithmic relationship between r_e and N_{100} from Papers II and III. The solid lines have a statistically significant correlation and the dotted lines are the 95 % confidence intervals of the relationships.

In Paper III the effects from meteorological parameters such as RH, SH, LTSS and CTT on r_e were investigated. However, none of these parameters were found to have a stronger correlation with the r_e than N_{130}. The RH in the boundary layer, though, was significantly correlated to r_e at both Vavihill and Hyytiälä.

4.2.2 Cloud optical thickness

The results regarding COT did not show the same consensus between the studies. In Paper II, the comparison between polluted and clean pixels (Figure 10a) showed that the clouds formed in the polluted air masses generally had a higher COT than those formed in cleaner air masses. This agrees with the proposed first indirect aerosol effect. Nonetheless, the polluted air masses all originated south of Barents Sea (relatively warm land areas) while all the clean air masses originated north of the area (cold oceans areas). Hence, some of the differences in COT may be due to the different meteorological conditions of the air masses.

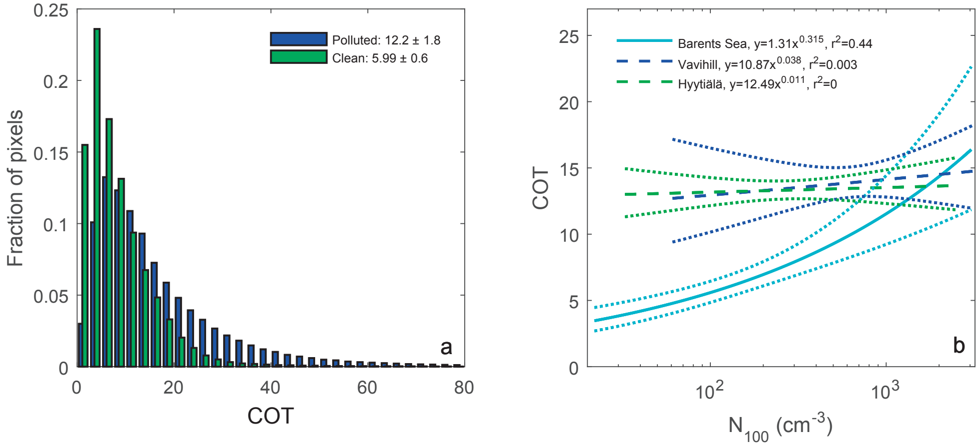

Figure 10
a) Normalised COT distributions of the polluted and clean pixels and geometrical averages of the distributions with corresponding uncertainties from Paper II. b) Logarithmic relationship between COT and N_{100} from Papers II and III. The solid line have a statistically significant correlation, the dashed lines have a non-significant correlation and the dotted lines are the 95 % confidence intervals of the relationships.

In Paper III, on the other hand, the COT was not found to vary with aerosol number concentration which can be seen in Figure 10b. As in Figure 9b, N_{100} is used in order to compare the results from Papers II and III. The marine clouds in Paper II had a lower COT than the continental clouds at low N_{100} values, but the COT increased to values similar to the continental clouds at higher N_{100}. In Paper III the meteorological parameters were not found to vary, to any great degree, with air mass origin. Different meteorological conditions in air masses of different N_{130} did therefore not seem to be responsible for the lack of correlation between COT and N_{130}. Instead, dynamical changes due to smaller droplets that cause changes in entrainment, may be the cause of the missing first indirect effect in the continental clouds.

4.2.3 Aerosol cloud interaction

The ACI (Aerosol cloud interaction) is a concept that was introduced to be able to compare aerosol cloud interaction results from different types of studies (Feingold et al., 2001; McComiskey and Feingold, 2008). The ACI is defined as:

$$ACI = \left.\frac{\partial \ln COT}{\partial \ln \alpha}\right|_{LWP} = \left.\frac{\partial \ln r_e}{\partial \ln \alpha}\right|_{LWP} = \frac{1}{3}\frac{\partial \ln N_d}{\partial \ln \alpha} \tag{4}$$

where α is proxy of CCN concentration. For the first two parts of the equation, the LWP needs to be constant in the calculations of the partial derivatives. In practice, the data are often divided into LWP bins and the partial derivatives calculated for each interval. This has been done in Paper III (Figure 11) but only for the r_e at 3.7 μm since COT is used in the calculation of the LWP. In Paper II, the N_d was calculated from the r_e and COT and hence the ACI could be calculated using part 3 of equation 4. Since part 3 of equation 4 does not require a division according to LWP, the results from Paper II are shown as a horizontal line in Figure 11.

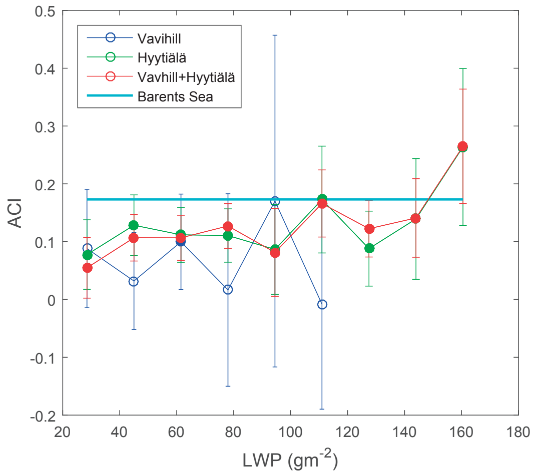

Figure 11
ACI as a function of LWP from Papers II and III. The r_e at 3.7 μm was used and α was set to N_{130} in the calculations of ACI for Vavihill Hyytiälä and both datasets together. For the Barents Sea, the N_d and N_{80} were used in the calculations of the ACI. The solid round markers show statistically significant correlations (at a 95% confidence interval) between r_e and N_{130} at that LWP interval.

The ACI at the Barents Sea is higher than the ACI in almost all LWP bins at the two continental sites, separately and combined. The ACI for Hyytiälä and combined dataset seem to increase slightly with LWP. However, the ACI from the highest LWP bins are most uncertain since these contain the least amount of data. The values of ACI found here are similar to those found in other remote sensing studies, which often have lower values than in-situ studies (McComiskey and Feingold, 2008).

4.2.4 Precipitation

In Paper III, precipitation data from the BALTEX project radar network were used to estimate whether aerosol number concentration affects the precipitation rate of the low-level clouds. The relationship between N_{130} and the average radar reflectance (dbzc) for the precipitating pixels is shown in Figure 12. Days in the study lacking precipitation were not included in the figure. For both Hyytiälä and Vavihill, higher aerosol number concentrations were associated with lower precipitation intensity. The correlation coefficients were significant both with (Figure 12a) and without (Figure 12b) logarithmic values of N_{130} for Hyytiälä. However, for Vavihill only the correlation in Figure 12b was significant. Previous studies (e.g. McComiskey et al., 2009) have found precipitating and none-precipitating clouds to respond differently to changes in aerosol loading. The data in Paper III were divided into precipitating and none-precipitating cases but the results regarding r_e and COT were not different for the different groups.

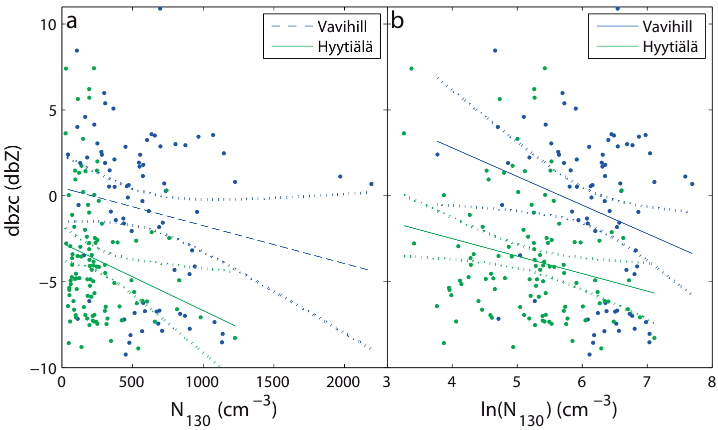

Figure 12
dbzc as a function of N_{130} for precipitating cases only. In b), the natural logarithm was applied to the N_{130}. The dotted lines represent the 95% prediction intervals for the relationships. The solid lines have a statistically significant correlation, the dashed line have a non-significant correlation and the dotted lines are the 95 % confidence intervals of the relationships (Sporre et al., 2014).

4.3 Convective clouds

Convective clouds over the Vavihill and Hyytiälä were the focus of Paper IV in which the profile method described in Section 2.2 was used. 9 (10) years of data were investigated for Vavihill (Hyytiälä) and the number of cases included in the study were 388 (295).

4.3.1 Effective radius

High aerosol number concentrations were found to be associated with low r_e at all levels of the clouds. The profiles were divided into 6 intervals according to N_{80} and a median profile was calculated for each interval (Figure 13). The profile displayed a rainbow-like appearance with the clouds formed in the air with the highest N_{80} having the smallest r_e. The profiles were also divided according to meteorological parameters such as cloud base temperature (T_B), CAPE, SH, RH and vertical velocity (w). T_B was found to affect the level at which the r_e starts growing rapidly with height. The temperatures at which this rapid increase occurs indicate that it is the mixed-phase zone (Section 2.2) and not the coalescence zone that is visible in the profiles. As can be expected, the level at which the clouds reach the mixed zone depend on the T_B. Profiles with low SH had low r_e but none of the other meteorological parameters separated the profiles visibly from each other.

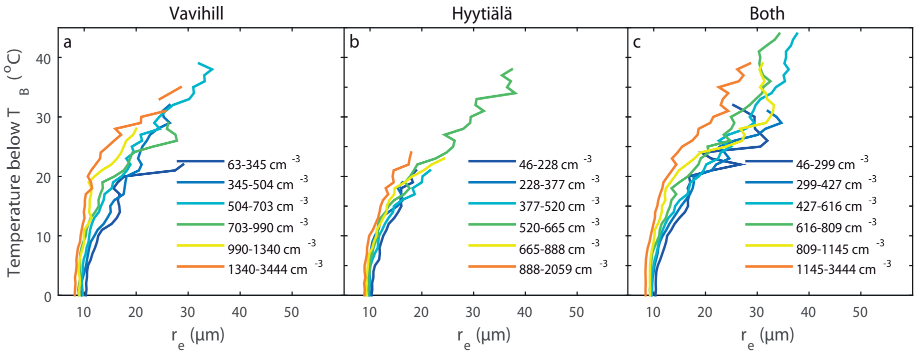

Figure 13
r_e profiles divided into 6 percentiles according to N_{80} for a) Vavihill, b) Hyytiälä, and c) both datasets together.

The COT profiles were divided according to the same parameters as the r_e profiles. However, none of the parameters separated the profiles significantly.

4.3.2 Precipitation

The BALTEX radar data and ground-based precipitation data were also used to study precipitation in convective clouds. Profiles producing more precipitation had both larger droplets and greater temperature difference between cloud base and cloud top (dT) than those producing little or no precipitation according to both types of precipitation datasets. Since the vertical extent of clouds affects their ability to produce precipitation, the convective cases were divided according to dT when examining the relationship between N_{80} and radar reflectivity (Figure 14). Most of the dT intervals show a significant decrease in dbzc with increasing aerosol load. The clouds with the highest dT have steeper slopes and hence seem more sensitive to aerosol perturbations.

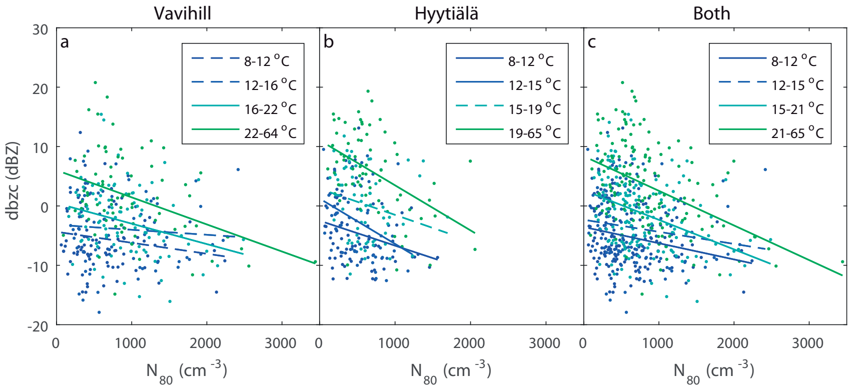

Figure 14
dbzc vs. N_{80} for a) Vavihill, b) Hyytiälä, and c) both datasets together. The data have been divided into 4 subsets according to dT. Solid lines have a statistically significant correlation with a 95 % confidence interval while profile with dashed lines have non-significant correlations.

4.4 Cirrus clouds

Cirrus clouds and how they are affected by aerosol particles transported down from the LSM was the topic of Paper V. Satellite remote sensing was combined with in-situ measurements of aerosol particles from the CARIBIC project in this study.

4.4.1 Volcanic influence on LMS and UT

Since 2005, a few explosive volcanic eruptions have increased the aerosol burden in the stratosphere (Vernier et al., 2011). The explosive volcanoes eject matter into the stratosphere where it is transported upwards in the tropics and then poleward. The air starts sinking at the midlatitudes and this downward transport is strongest in the spring. The transport from the tropics to the midlatitudes can, however, take from a few months up to several years. The levels of sulphate aerosol measured by the CARIBIC platform in the LMS and UT are shown in Figure 15. The sulphate mass concentration (S) values were divided by PV since there is a strong particulate sulphur gradient in the LMS (S concentrations increase from the tropopause into the LMS). The measurements during 1999-2002 show low sulphate particle levels, a period with low volcanic impact on the stratosphere. After 2005, the LMS sulphate levels increased, especially after the eruptions of the extra-tropical volcanoes *Kasatochi* in 2008 and *Sarychev* in 2009 as well as the tropical volcano *Nabro* in 2011. However, the influence of fresh volcanic plumes on the UT/LMS region was not the focus of Paper V, but rather how the change in background stratospheric sulphate concentrations affected the UT at midlatitudes. Consequently, the data directly influenced by major eruptions have been excluded from the analysis.

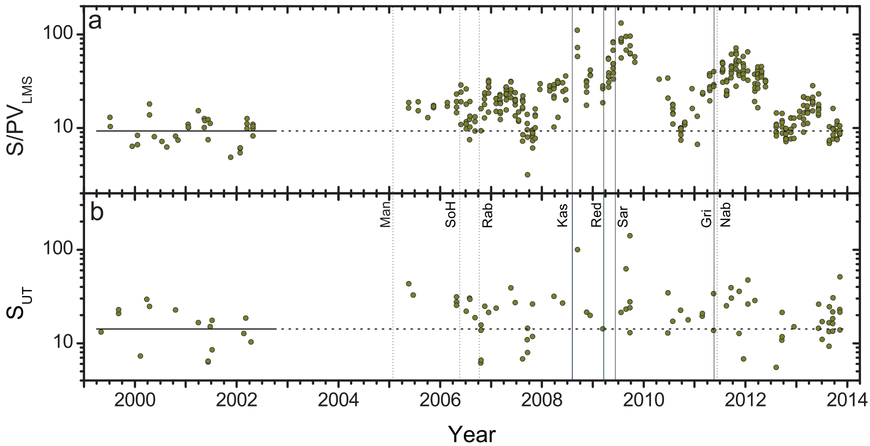

Figure 15
Temporal variations in the northern midlatitude of a) S/PV values in the LMS (S/PV_{LMS}; ngm^{-3} STP/PVU), and b) sulphur concentrations in the UT (S_{UT}; ng m^{-3} STP). The horizontal lines show geometric averages for the period 1999-2002. Vertical lines mark volcanic eruptions identified to have influenced the particulate sulphur concentrations in the LMS. Vertical dashed and solid lines represent eruptions in the tropics and northern midlatitudes, respectively. Data for the 30 days following these eruptions are excluded (Friberg et al., 2015).

4.4.2 Sulphate aerosol influence on cirrus clouds

To investigate whether cirrus clouds at midlatitudes are affected by volcanic aerosol transported into the UT from the LMS, the CARIBIC data were combined with the MODIS Level 3 CR products. Data from March to July were used since this is the time of the year when the subsidence from the stratosphere is the strongest. Figure 16a shows the variation of the average of CR and LMS sulphate concentrations from 2000 to 2014. Some years are missing in the CARIBIC dataset either due to no measurements being made or too few months of CARIBIC data being available. The Terra and Aqua datasets co-vary nicely but the Aqua data are approximately 20% higher. This is due to the later overpass of this satellite, which allows for the influence of more convective activity that creates anvils. Since anvils are expected to be less sensitive to changes in downwelling stratospheric aerosol, only the Terra data were further investigated. The year-to-year variation in CR resembled the variation in S/PV well (note that the axis of the sulphate data is reversed) and the anti-correlation of the S/PV and Terra CR can also be seen in Figure 16b. The CR decreased 8±2% between 2001 and 2011 which is associated with a LMS particulate sulphate concentration increase by a factor of 3-4.

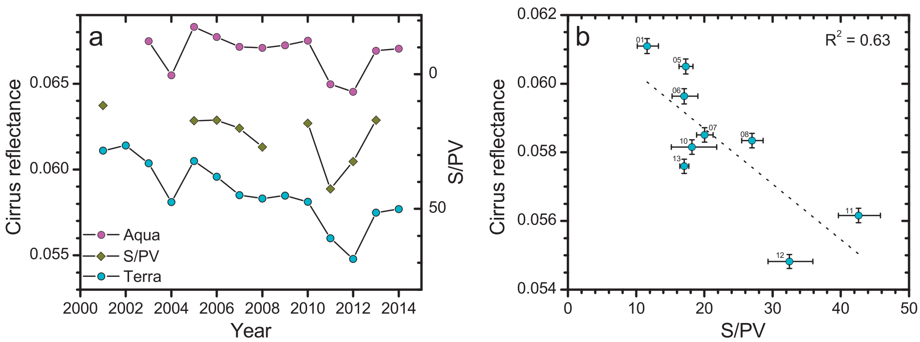

Figure 16
a) Time series of the geometric average S/PV in the LMS (S/PV_{LMS}; ngm^{-3} STP/PVU) (note the reversed scale) and the arithmetic average of the MODIS retrieved parameter, CR from Aqua and Terra, for samples taken below 5 PVU in the season March-July. b) A comparison of S/PV and Terra's CR as in a). A linear regression fit is shown as a dotted line (Friberg et al., 2015).

In Figure 17, the average CR from 2 years of low volcanic activity (2001 and 2002) is compared to the average CR from 2 years with the highest volcanic impact (2011 and 2012). The areas with the largest decrease in CR are found over the mid-Pacific, North American continent, the Atlantic Ocean and Europe.

Possible causes for the decrease in CR due to an increase in sulphurous aerosol in the UT are:

- Increased sulphurous aerosol concentrations in the UT and LMS can increase the temperature and decrease vertical velocities. In a homogeneous cirrus formation regime this results in a reduced ice crystal formation rate, which leads to optically thinner clouds (Kuebbeler et al., 2012).
- The downwelling sulphate aerosol can act as IN and shift the nucleation regime from homogeneous to heterogeneous which results in thinner clouds.
- Sulphuric acid aerosol could coat and deactivate existing IN, which in a heterogeneous nucleation regime would decrease the CR.

Optically thinner clouds both reflect less solar radiation and absorb less longwave radiation. The latter effect, however, is dominant and the 8% decrease in CR thus implies a negative RF at the midlatitudes in the Northern hemisphere.

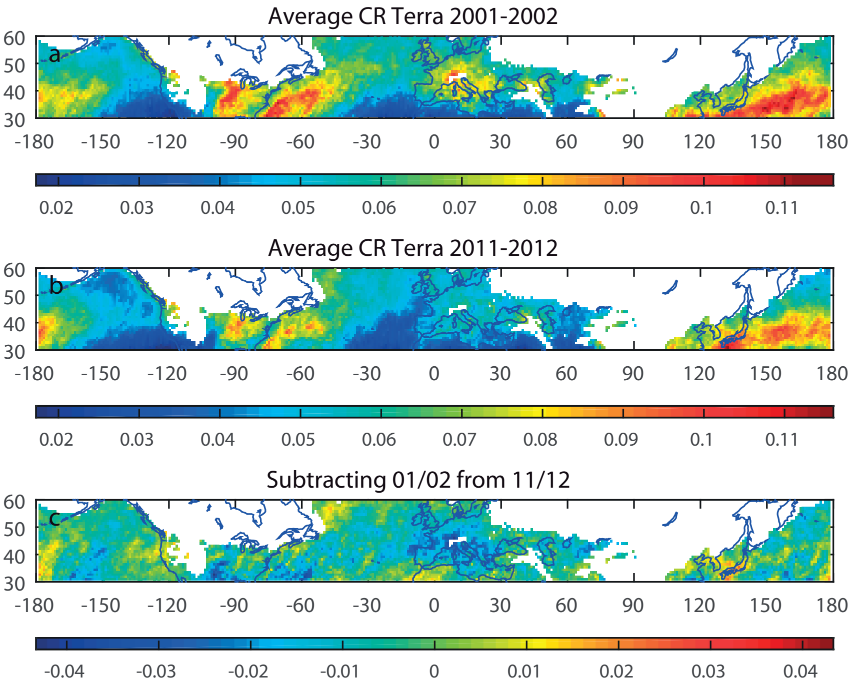

Figure 17
Geographical distributions of the March-July CR averaged over the years of a) background concentrations of sulphur (2001-2002), b) years of strongest influence of volcanism (2011-2012), and c) the difference in CR between the years in Figures 17b and 17a (Friberg et al., 2015).

5 Conclusions and Outlook

In the research presented in this thesis, satellite measurements of clouds have been combined with a range of other datasets to investigate aerosol-cloud-precipitation interactions. These are the main conclusions of the research:

The comparison of ground-based remote sensing to MODIS satellite data shows a relatively good agreement between the datasets. Nevertheless, the MODIS data overestimate the CTH for single layer clouds and underestimate the CTH when also multilayer clouds are included in the comparison. The median differences are relatively low but there are cases where the difference is several thousand meters. The LWP retrievals agree very well between the satellite and ground-based measurements up to 200 gm^{-2}, above which the differences become much larger. It is reassuring that the MODIS LWP values are similar to their ground-based counterparts for most parts of the dataset, especially since the MODIS LWP values were not retrieved directly but calculated from the r_e and COT.

By combining satellite cloud data and in-situ aerosol measurements, it was found that the cloud droplet sizes, represented by r_e, decreased with increasing aerosol number concentrations. This was true in low-level clouds over the ocean, low-level clouds over land and convective clouds over land. How the clouds were affected by meteorological conditions such as temperature, RH, SH and atmospheric stability was also investigated but none were found to have a stronger relationship with the r_e than the aerosols.

The results are not as homogeneous for the COT. For the investigated low-level clouds over the ocean, the COT increased with increasing aerosol loading. However, it cannot be ruled out that this was partly caused by different meteorological conditions of the polluted and clean air masses. For the low-level and convective clouds over land, no change in COT was observed when the aerosol number concentration varied. Changes in cloud dynamics due to smaller droplet sizes is a plausible cause for the missing effect of aerosol particles on COT in the clouds over land. One can conclude that the first indirect aerosol effect was not present in all clouds investigated in the thesis.

Weather radar data were used to study the precipitation intensity of the investigated clouds. The intensity of the precipitation was associated with a weak decrease as the aerosol number concentration increased in both low-level and convective clouds over land. As the convective clouds with higher cloud tops are more prone to precipitate, the convective clouds had to be sorted according to vertical extent of the clouds for the signal to appear.

Aerosol data from the CARIBIC project were combined with satellite retrievals of cirrus clouds. The CR at midlatitudes in the northern hemisphere was found to decrease when the downwelling air from the LMS contained higher levels of sulphate aerosol. Because cirrus clouds have a net warming effect on the climate, lower CR leads to a cooling. The variations in the sulphate concentrations are a result of the eruptions of explosive volcanoes. It thus appears that volcanoes can affect the climate through alterations of cirrus cloud properties.

The research on how microphysical and optical properties of clouds change with aerosol number concentration presented in this thesis adds information to the ongoing research regarding the first and second indirect aerosol effects. The research contributes to a reduction in the uncertainties associated with aerosol-cloud-precipitation interactions by providing information on these processes in a region with relatively low aerosol number concentrations and quite weak dynamical meteorological forcing. The thesis also presents the first observational study of long-term volcanic influence on cirrus clouds.

The research on aerosol-cloud-precipitation is nowhere near finished. There are still large uncertainties regarding how much aerosols affect climate through interactions with clouds. The uncertainties range from the microscale (droplet activation) to the mesoscale (single cloud dynamics and cloud cluster dynamics) to the synoptic scale (How do large scale changes in aerosol burden affect synoptic scale cloud systems?). The research presented in this thesis contributes to information on the mesoscale but more research is needed on all scales.

Evidence of the first indirect aerosol effect is missing in some clouds investigated in this thesis. This could be due to the changes in cloud dynamics caused by smaller droplets that increase the entrainment of air into the clouds. If this is the cause, and in which clouds the indirect aerosol effect is absent, together make up an important topic for future studies to better understand to what degree aerosol loading affects clouds. Both observational and modelling studies are required to tackle this problem.

One cloud type still associated with particularly large uncertainties is the cirrus cloud. Whether it is homogeneous or heterogeneous ice nucleation mechanisms that dominate cirrus cloud formation is one topic that needs more research. More research into how cirrus clouds are affected by particle levels in subsiding stratospheric air is also needed. This topic is especially important with regards to the ongoing geoengineering discussions, as one of the proposed geoengineering methods is to emit sulphate particles into the stratosphere.

The research presented in this thesis is almost exclusively based on large, freely available datasets. It is vital that such datasets continue to be free of charge and easily accessible to enable similar types of research in the future.

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