M.Sc. Thesis
Meteorology

Source areas and effect of wet deposition on particles detected at SMEAR II measurement station as studied with FLEXPART

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Tiivistelmä — Referat — Abstract

Aerosol particles are one of the most studied phenomena of modern days because their concretion and chemical compositions affect, both directly and indirectly, many aspects of society, for example, human health. The source areas of the particles affects their chemical composition and concentration and thus it is important to know where aerosols originate. In addition to the source areas of particles, it is important to understand the processes that affect the particles during transport through the atmosphere, for example, wet deposition. The aims of the thesis were to find out the characteristics in the source areas (1) and the effect of wet deposition (2) on the particles detected at SMEAR II (Hyytiälä, Finland) measurement station. Both aims were studied with the FLEXPART model and to study the first aim also the Hysplit model was used. FLEXPART is a Lagrangian particle dispersion model and Hysplit is traditional trajectory model. The both aims were studied with 14 case studies and each case study was studied with 96 hour backward in time simulations. The Hysplit simulations were made for each case study once and the FLEXPART simulations were made for each case study three times.

The 14 case studies were selected based on observation made at the measurement station SMEAR II during the time period between February and April 2014. The case studies were divided into two classes: cloud and aerosol cases and the general characteristics of the cloud and aerosol cases were compared to each other. The extent and location of the source areas between FLEXPART and Hysplit were compared to define to source areas of the particles. The effect of wet deposition in the FLEXPART simulations was studied by comparing two simulation types: with default and with modified wet deposition parameterizations.

The results of the thesis suggest that the measurement altitude, i.e. the release altitude in backward in time simulation, has a significant effect on the source areas of particles. The results also indicates that in cloud cases the particles originate more wider area than in aerosol cases. In cloud cases most of the trajectories come from South or South-West whereas in the aerosol cases most of the trajectories come from North or North-West which indicates that the source area of the particles could be in the background causing different interesting observations. The results suggest that wet deposition has a major role both in the amount of the mass and in the source areas of the particles detected in SMEAR II (Hyytiälä, Finland), especially in low pressure and cyclone situations, i.e. in cloud cases. Also the result indicates that the amount of scavenged mass is much higher in-cloud than below-cloud.

Avainsanat — Nyckelord — Keywords

FLEXPART, Particle, Dispersion Model, Trajectory, Wet Deposition, SMEAR II

Säilytyspaikka — Förvaringsställe — Where deposited

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Tiivistelmä — Referat — Abstract

Aerosolipartikkelit ovat yksi nykypäivän tutkituimpia tutkimusaiheita ilmakehätieteiden saralla, koska niiden pitoisuus ja koostumus vaikuttavat moniin ihmiselämän osa-alueisiin, kuten terveyteen, sekä suoraan että välillisesti. Aerosolien lähdealueet vaikuttavat niiden kemiallisen koostumukseen ja pitoisuuteen, minkä takia partikkeleiden lähdealueiden ja niihin vaikuttavien tekijöiden tunteminen on tärkeää. Lähdealuieden lisäksi myös ilmakehässä tapahtuvien, partikkeleiden kuljetukseen liittyvien, prosessien tunteminen on tärkeää. Yksi tälläinen prosessi on esimerkiksi partikkeleiden märkälaskeuma. Tämän tutkielman tavoitteena oli tutkia partikkeleiden lähdealueiden yleispiirteitä (1) sekä märkälaskeuman vaikutustusta SMEAR II mittausasemalla (Hyytiälä, Suomi) havaittuihin partikkeleihin. Tavoitteiden tutkimuseen käytettiin FLEXPART ja Hysplit malleja. FLEXPART on partikkeleiden leviämistä kuvaava malli ja Hysplit on perinteinen trajektorimalli. Tavoitteita tutkitiin tekemällä 96 tunnin pituisia ajassa taaksepäin laskettuja simulaatioita 14 eri tapaustutkimukselle. Jokainen tapaustutkimus simuloitiin kerran Hysplitillä ja kolmesti FLEXPART:lla.

Tutkitut 14 tapaustutkimusta valittiin SMEAR II asemalla helmi-huhtikuussa 2014 tehtyjen mielenkiintoisten havaintojen perusteella. Tapaustutkimukseen jaoteltiin pilvitapauksiin ja aerosolitapauksiin, minkä jälkeen näiden kahden eri luokan yleispiirteitä vertailtiin keskenään. Partikkeleiden lähdealueiden määrittämiseksi FLEXPAR ja Hysplit simulaatioiden mallintamia partikkeleiden lähdealueiden laajuuksia ja sijainteja vertailtiin toisiinsa. Märkälaskeuman vaikutuksen tutkimiseksi simulaatioissa käytettiin kahta eri märkälaskeuman parametrisointitapaa: oletustapaa sekä muunneltua tapaa.

Tämän tutkimuksen tulokset esittävät, että partikkeleiden mittauskorkeudella, eli takaisinpäin ajassa tehtävän simulaation kannalta tarkasteltuna vapautuskorkeudella, on huomattava vaikutus partikkeleiden lähdealueiseen. Tuloksesten perusteella pilvitapauksissa partikkeleiden lähdealue on paljon laajempi kuin aerosolitapauksissa. Pilvitapauksissa suurin osa trajektoreista kulki SMEAR II etelään tai lounaaseen, kun taas aerosoltapauksissa suurin osa trajektoreista kulki asemalta katsottuna pohjoiseen tai luoteeseen. Tämä viittaa siihen, että partikkeleiden lähdealueella voi olla vaikutusta pilvi- ja aerosolitapausten kaltaisiin havaintoihin. Tulokset esittävät, että märkälaskeumalla on huomattava vaikutus SMEAR II asemalla havaittaviin partikkeleihin erityisesti matalapainetilanteissa, joita havaittiin pilvitapausten aikana. Tulokset myös osoittavat märkälaskeuman olevan suurempaa pilven sisällä kuin sen alapuolella.

Avainsanat — Nyckelord — Keywords

FLEXPART, Partikkeli, Leviämismallinnus, Trajektori, Märkälaskeuma, SMEAR II

Säilytyspaikka — Förvaringsställe — Where deposited

Kumpulan tiedekirjasto

Muita tietoja — övriga uppgifter — Additional information

Contents

1 Introduction

One of the most studied phenomena of modern days is aerosol particles. Aerosol particles are collections of solid or liquid particles suspended in gas (Hinds, 1999). Aerosol particles are an object of interest because their concentration and chemical compositions affect many aspects of society, for example human health (Poschl, 2015) and agriculture through cloud formation (Roscoe et al., 2015). Sources of aerosols can affect their chemical compositions and concentrations (Mengfei et al., 2015) and thus it is important to know where aerosols originate. The traditional method to identify the source regions of aerosol is to calculate backward trajectories. Backward trajectories describe the path of the particle backward in time (Holton et al., 2013). With backward trajectories there are a few limitations, for example subgrib scale processes can not be accounted for in traditional backward trajectory models and the error in backward trajectory increases linearly with increasing distance along the paths of trajectory (Kahl,1993). One way to avoid these limitations and try to reduce the error is to use Lagrangian particle dispersion models to identify the source regions of particles. Lagrangian particle dispersion models can simulate physical processes and motions of individual particles and thus are useful when studying aerosol particles.

In Langrangian particle simulations, in addition to advection, a wide range of atmospheric processes have to be, and can be, simulated because those processes affect motions and concentration of particles. These processes are, for example, turbulence, convection, diffusion and wet deposition. On this thesis wet deposition process is under research. Wet deposition is the removal of particles from the atmosphere to the Earth's surface by atmospheric hydrometers, for example, by cloud droplets or ice crystals (Seinfeld et al., 2006). Wet deposition can take place both in-cloud and below-cloud.

One widely used and freely available Lagrangian particle dispersion model is FLEXible PARTicle dispersion model (FLEXPART). Most previous studies which have utilized FLEXPART used it in forward mode, when the transport of particles is under research (Marzo, 2014). FLEXPART can also be used in backward mode, when interest is in the potential sources of particles (Jordan et al., 2015; Gadhavi et al., 2015). Only a few of these FLEXPART studies which used it in backward mode have been focused on the effect of wet deposition.

In this thesis both source regions of particles and the effect of wet deposition at measurement station SMEAR II (Hyytiälä, Finland) are under research. These themes are studied with 14 case studies from spring 2014. The case studies are 7 new particle formation cases (called from now on aerosol cases) and 7 interesting cloud case studies (called from now on cloud cases). The main tool for research is FLEXPART, but also HYbrid Single Particle Lagrangian Integrated Trajectory (Hysplit) model is used. Hysplit is traditional backward trajectory model.

There are two aims on this thesis: first aim is to find out how FLEXPART can simulate the source regions of particles during the case studies and what differences there are in source regions between cloud and aerosol cases. To study the first aim trajectories of FLEXPART and Hysplit simulations are compared to each other. The second aim is to study the impact of wet deposition on how much mass is transported to SMEAR II (Hyytiälä, Finland). The second aim is studied by regarding the mass of the particles originating from the simulations. Also the differences between in-cloud scavenging and below-cloud scavenging are studied during the second aim.

The structure of the thesis is as follows. After the Introduction in Chapter 2, the theory behind Lagrangian particle dispersion models is explained, also FLEXPART and Hysplit models and theories behind them are introduced. In Chapter 3 output data and data processing is discussed. In Chapters 4 and 5 results from trajectory and wet deposition studies are presented. Chapter 6 is discussion and 7 is summary.

2 Theory and background

According to Holton and Hakim (2013) all atmospheric motions are governed by conservation of mass, momentum and energy. These conservations of mass, momentum and energy can be handled mathematically by two different ways, with a Eulerian method or with a Lagrangian method. In both methods all of the calculations are done for infinitesimal control volume of fluid, in atmospheric cases the fluid is air. This infinitesimal control volume of air is commonly referred to as air parcel.

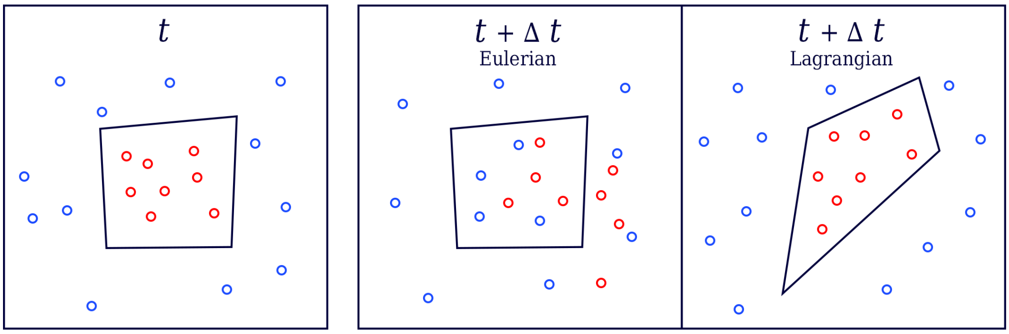

Figure 1: *Schematic figure of the differences between Eulerian and Lagrangian calculation methods after a time step Δt. The square marked with solid black line represents the air parcel.*

Figure 1 shows schematically the difference between Lagrangian and Eulerian methods. The air parcel contains at initial time t all of the red particles (left panel) which describe the mass inside the air parcel at initial time t. In the Eulerian method the position of the air parcel is fixed relative to the coordinate axes and the shape or volume of parcel does not change after a time step Δt but mass, momentum and energy can flow through the air parcel (middle panel). In other words, in the Eulerian method, the calculation grid and output grids are the same, both can be for example expressed in geographic coordinates. In the Lagrangian method the air parcel follows the motions of flow and consists of all of the same particles after a time step Δt than in the initial time t, i. e. the mass of the air parcel remains constant (right panel). In the Lagrangian method the calculations are made in a different grid from the output grid. The calculation grid consists of an individual air parcel which follow the flow and the motions are relative to the initial point while the output grid can be for example converted to geographic coordinates.

Although all calculations are based on either Lagrangian or Eulerian calculation methods there are different variations on how these calculation methods are used in atmospheric models. Models can be either fully Eulerian or fully Lagrangian, or hybrid model between Eulerian and Lagrangian methods. Two model types that are based on these Lagrangian and Eulerian calculations methods are trajectory models and Lagrangian particle dispersion models.

Trajectory models calculates trajectories. A trajectory is defined to be a curve that describes the path of a specific air parcel followed over a finite period of time (Holton et al., 2013). The problem with trajectory models is that they are highly uncertain and very sensitive to initial values. One of the biggest uncertainties with trajectory models is starting position error, which is directly dependent on the starting point of the trajectory. To reduce the effect of starting position error, developing of ensemble trajectories started in the 1980's (Stohl, 2002). Ensemble trajectories are calculated by shifting the initial position of the air parcel relative to the meteorological grid to some new position in the horizontal or vertical direction and then advection and dispersion calculations are done in this new position. After advection and dispersion are calculated, the air parcel is adjusted back to the main initial position and the motions of air parcel on a time step are calculated based on the advection and dispersion calculated elsewhere (Draxler, 2003). By differing the initial position slightly in vertical and horizontal directions multiple times, multiple trajectories, called ensemble trajectories, are calculated. Ensemble trajectories are still singly uncertain but with the other ensemble trajectories they give estimations of the starting position error. In addition to the starting position the error of trajectories depends on for example the calculation methods and the meteorological input data. Stohl (1998) brought together different trajectory studies and suggested that generally the error in trajectories is 20% of the length of the path that the air parcel has gone. The reason why trajectory models are commonly used despite their errors is that they are quick and effective models and their applications can vary between synoptic meteorology situations and climatology.

Lagrangian particle dispersion models (LPDM) are based on the Lagrangian method and LPDM are basically extended from traditional trajectory models and their base and the structure may be based on some existing trajectory models (Stohl, 2002). Particles themselves have a Lagrangian nature since they move by the flow and thus it is natural to use Lagrangian methods while calculating the movements of particles (Zannetti, 1992). In addition, Lagrangian models do not have numerical diffusion like Eulerian models, which also promotes the usage of Lagrangian method (Nquyen et al., 1997). The idea of LPDM is to use a certain number of particles to simulate the flow and dynamic processes in the atmosphere (Stohl et al., 2005). These simulated particles are released at a specific point or volume and their dispersion with the flow is calculated either forward or backward in time. LPDMs are good for simulating physical processes that are usually subgrid scale. For example turbulence in the boundary layer is a subgrid scale process that can be represented on a some level in LPDM. Traditional trajectory models can not describe turbulence because one or even a few dozen trajectories are not enough to represent turbulent mixing (Stohl, 2002). Also LPDMs are good tools to describe complex physical and chemical processes in the atmosphere (Stohl et al., 2005). Like every atmospheric models also LPDMs have some limitations. The main limitation with LPDM is associated with the turbulence, even though the turbulence is a process that can be described better in LPDM than in traditional trajectory models. The problem with turbulence in LPDM is the determination of the turbulent velocities which describes the diffusion of the particles in the boundary layer (Stohl, 2002).

In this thesis two models are used to study the aims of the thesis. The main model is the Lagrangian particle dispersion model FLEXPART and the second model is the trajectory model Hysplit. In the following chapters 2.1 and 2.2 both models and the theories behind them are introduced.

2.1 FLEXPART introduction

FLEXPART simulates long-range and mesoscale transport together with all linear physical processes like diffusion, and also if the user so wishes, radioactive decay and wet and dry deposition (Stohl et al., 2005). The only processes that FLEXPART can not simulate are all non-linear chemical reactions (Stohl, 2002). FLEXPART can be run either forward in time or backward in time. Forward in time FLEXPART simulates dispersion of tracer particles from their sources downwind on the calculation grid and backward in time it simulates potential sources for particles detected in specific locations. In forward in time simulation the release area of particles is the source of particles and in backward in time simulation the release area is the detection point. In addition to particles also tracer gases can be used as tracer in FLEXPART. The deposition processes and calculation methods of gases differ from the processes and calculations of particles and thereby the theories and formulations showed below only apply to particles not gases.

FLEXPART is based on the kinematic trajectory model FLEXTRA and it's developing process started in 1998 (Stohl et al., 2005).

Development has been continued to present day and the newest version is FLEXPART version 9.02, which is used in the simulations of this thesis. Version 9.0 was released in June 2012. FLEXPART is under development all the time and the number of FLEXPART users is growing. Different versions with new parameterizations are evaluated with different experiments, for example, version 2.0 was evaluated with tracer experiments (Stohl et al., 1998) and version 4.0 with air pollution studies (Stohl and Trickl, 1999; Forster et al., 2001; Spichtinger et al., 2001; Stohl et al., 2002, 2003). With these evaluation studies it has been proved that FLEXPART is capable of describing particles' dispersion and different physical processes associated with particles' motions and concentrations.

One of the good points in FLEXPART, and potentially one reason why the number of users is growing fast, is that how freely the user can set all variables of the simulation. For example, almost all physical parameters, calculation areas, release times and particle distributions are user definable. These variables are set in FLEXPART's options where the users can also define which physical processes, for example convection and sub grid parameterization, are on or off during the simulation. The number of released particles is one of the parameters which are defined in options. The number of particles has to be large when simulating dispersion of particles. Large number of particles means in this case tens or hundreds of thousands of released particles. One reason why the number of particles has to be large is that if there are small amount of particles, concentrations are reduced very quickly when the particles spread over the atmosphere and the differences comes too small to detect. The second reason why the number or particles has to be large is associated with the clustering process (Chapter 2.1.3).

In first FLEXPART versions it was possible to use meteorological input data only from numerical prediction model of the European Centre for Medium-Range Weather Forecasts (ECMWF). At least since version 8.2 (Stohl et al., 2010) it has been possible to also use National Weather Service's National Centers for Environmental Prediction's (NCEP) Climate Forecast System Reanalysis version 2 (CFSR) data as input to FLEXPART. CFSR data is used as meteorological input data in the FLEXPART simulations of this thesis because of CFSR data is easier to use, based on that CFSR data is easier to access compared to ECMWF data and CFSR data has already been post-processed to be in the correct format for FLEXPART whereas ECMWF data has not. CFSR data are freely download at CISL Research Data Archive (UCAR, 2016).

CFSR is a coupled atmosphere-ocean-land surface-ice sea system reanalysis model. The model has been evaluated to be very accurate and it can describe seasonal fluctuations well (Saha et al., 2014). The model output is available every 6 hours, horizontal resolution is 0,5 degrees and there are 64 vertical layers in the data. There are multiple fields that FLEXPART requires from CFS data for its simulations: dewpoint temperature, geopotential height, land cover, planetary boundary layer height, pressure, mean sea level pressure relative humidity, temperature, u- and v-components of wind, vertical velocity and water equivalent of accumulated snow depth fields. To calculate the effect of wet deposition FLEXPART requires also cloud cover, large-scale and convective precipitation.

2.1.1 Backward in time simulations with FLEXPART

In FLEXPART backward in time simulations particles are released in a source or source areas and the signs of wind and other physical processes are negative and the model calculates where the particles came to the source. In backward in time simulations the main output of FLEXPART is source-receptor relationship. Source-receptor relationships describe sensitivity from receptor to source (Seibert and Frank 2004). The source-receptor relationship is higher in grid cells where more material is coming to source point and zero where there nothing is coming from. Source-receptor relationship output is not used in the results of this thesis and therefore it will not been explained in more detail in here. For further information about source-receptor relationship see Seibert and Frank (2004). In addition to source-receptor relationship there can also be a particle output and a trajectory output in the FLEXPART backward in time simulations. Particle output describes the properties of each particle, for example mass and location of the particle, at each time step. Trajectory output contains locations of plume trajectories and mean trajectory (Chapter 2.1.3) at each time step . More information about FLEXPART's outputs and handling of them for the analysis presented in this thesis is described in chapter 3.2.2.

Because all of the physical and chemical processes that FLEX-

PART can simulate are linear, the backward in time modeling is simple (Stohl, 2002). Linear physical processes can be calculated in backward in time simulations even if the idea of some of these physical processes, like wet deposition, is hard to understand backward in time. In a mathematical way, modeling of wet deposition is easy and it can be handled by changing calculation methods slightly (Seibert and Frank, 2004). For further information about backward in time modeling see Seibert and Frank (2004).

2.1.2 FLEXPART's wet deposition parameterizations

There are two main mechanisms to remove particles from the atmosphere, wet and dry deposition. Dry deposition is not considered at all in here, because only wet deposition is under research in the thesis. Wet deposition is a process which removes particles from the atmosphere by atmospheric hydrometers, like cloud droplets or ice crystals, and dry deposition takes into account all other processes, for example, sedimentation and impaction (Hinds, 1999). Wet deposition is the dominant removal process in regions of precipitation. For example, in Finland the mean annual precipitation as measured between 1981-2010 in Southern Finland is 700 to 750 millimeters and in Northern Finland it is from 450 to 500 millimeters (FMI Annual Statistics, 2010). Because Finland is clearly, based on the mean annual precipitation, an area where precipitation occurs, it is important to be able to understand and simulate the wet deposition process as well as simulating the movements of particles in Finland in situations where precipitation occurs. Wet deposition depends on the precipitation rate and the properties of the scavenged material.

Wet deposition occurs both in clouds (in-cloud scavenging) and below clouds (below-cloud scavenging). In-cloud scavenging aerosols are removed from the air by the nucleation process, where the particles act as condensation nuclei (Webster et al., 2014), or by impaction with hydrometers (Croft et al., 2010). Below cloud particles are swept out by falling hydrometers. Particles with diameter from 0,08 micrometers to about one micrometer can act as condensation nuclei when they increase in-cloud scavenging. Scavenging by impaction with hydrometer is effective for very small particles with the diameter smaller than 0,01 micrometers and for very big particles with diameter larger than a few micrometers. For the very small particles impaction is effective because they undergo Brownian diffusion and for big particles it is effective because they have larger inertial effects (Webster et al., 2014).

On the basis of Hertel et al. (1995) and Webster et al. (2014) the contribution of in-cloud scavenging is in most of the cases larger than the contribution of below-cloud scavenging. There are three main reason for this. To understand the first two reasons the terms below-cloud and in-cloud have to be regarded from a modeling point of view. Hertel et. al. (1995) assumed that below-cloud scavenging takes place below 250 meters, while in-cloud scavenging takes place in a layer between 250 meters and 2 kilometers. In FLEXPART these altitudes are calculated based on meteorological input data, but they are both in the lower part of atmosphere. Based on these altitudes in-cloud scavenging has larger contribution because firstly, the depth of cloud layer is much larger than the depth of the below-cloud layer. Hence there are more particles in the in-cloud layer than below-cloud, even if the particles' number concentration is larger near the surface than the in-cloud layer. Secondly the particles spend a longer time in-cloud than below-cloud as the in-cloud layer is deeper and the fall speed is the same in both layers. Also in cloud there are particles that do not fall out from the cloud, for example cloud particles. Thirdly the number concentration of particles is focused to particles with diameter less than one micrometer (Hinds, 1999) and in-cloud scavenging processes affects mostly to particles with diameters less than one micrometer. This causes that there are more processes that remove particles in-cloud than below-cloud. According to Hertel et al. (1995) below-cloud scavenging can be dominant only when the concentrations of the air is much larger in the below-cloud layer compared to the concentrations of the air in-cloud. Events where below-cloud scavenging has larger impact are really rare because below-cloud scavenging is valid only in the events with really heavy precipitation (Hertel et al., 1995).

FLEXPART uses humidity and temperature from meteorological input data to calculate the occurrence of clouds and if the relative humidity is over 80% cloud is assumed. For in-cloud scavenging FLEXPART uses scheme of Hertel et al. (1995) and below-cloud scavenging is handled with McMahon's (1979) exponential decay parameterization. In the both parameterizations wet deposition affects the mass of the particle by scavenging coefficients (in-cloud scavenging coefficient Λ_i and below-cloud scavenging coefficient Λ_b). The mass of the particle after a time step can be calculated by

$$m(t + \Delta t) = m(t) \exp(-\Lambda \Delta t) \tag{2.1}$$

where Λ is in-cloud or below-cloud scavenging coefficient and Δt is the magnitude of the time step.

The in-cloud scavenging coefficient Λ_i [s^{-1}] is calculated by

$$\Lambda_i = \frac{S_i I}{H_i}. \tag{2.2}$$

Equation 2.2 shows that in-cloud scavenging depends on precipitation rate I and the height over which in-cloud scavenging takes places H_i. H_i can be constant but in FLEXPART it changes between grid cells and it is set to respond to the calculated cloud depth. S_i is the in-cloud scavenging ratio which describes the ratio between concentration in precipitation and concentration in air in a grid cell. S_i depends on liquid water content cl which in turn depends on precipitation rate I. S_i is calculated by

$$S_i = \frac{0.9}{cl} \tag{2.3}$$

where the liquid water content cl is

$$cl = 2 \times 10^{-7} I^{0.36}. \tag{2.4}$$

The below-cloud scavenging coefficient Λ_b [s$^-$1] is calculated by

$$\Lambda_b = A I^B \tag{2.5}$$

where A is the scavenging coefficient, B is the below-cloud scavenging parameter and I is precipitation rate. A and B are species specific constants.

Precipitation rate I used in scavenging coefficients calculations is subgrid scale precipitation I_s. Subgrid scale precipitation takes into account subgrid variability of the model-simulated rain. Subgrid variability has to be taken into account because wet deposition depends non-linearly on the precipitation rate. I_s is calculated by

$$I_s = \frac{I_l + I_c}{F} \tag{2.6}$$

where I_l is large scale precipitation, I_s is convective precipitation and F area fraction of grid cell which experiences precipitation. F is calculated by

$$F = \max\left[0.05, CC\frac{I_l fr_l(I_l) + I_c fr_c(I_c)}{I_l + I_c}\right] \tag{2.7}$$

where CC is total cloud cover at the grid cell, I_l is large scale precipitation, I_c is convective precipitation and fr_l and fr_c are correction factors that depend on the precipitation rates of the meteorological input data and they describe the non-linear dependence of the wet deposition and precipitation rate. Dependencies between precipitation rates and correction factors are listed in Appendix A.

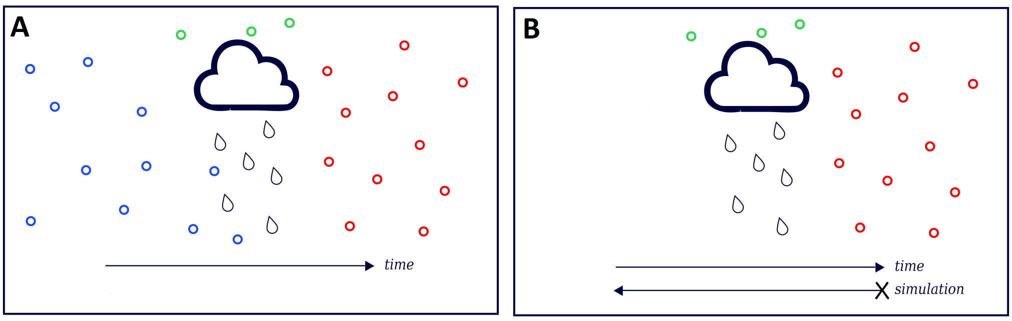

Figure 2: *Schematic figure from wet deposition* ***a.*** *in nature, what physically happens,* ***b.*** *in a FLEXPART simulation, what happens in the simulation . Figures show the situation in one grid cell.*

In the forward in time simulation the effect of wet deposition is the same as what physically happens in nature: particles move with the wind and when clouds and rain occur, they remove the mass of the particles by wet deposition. In the backward in time simulation the effect of wet deposition is not the same as what physically happens and because of that wet deposition in backward in time simulation is hard to understand. To explain what happens, Fig. 2a shows schematically what happens physically when wet deposition occurs and Fig. 2b shows what happens in FLEXPART backward in time simulation when wet deposition occurs. Both figures describe the situation in one grid cell, which is in this schematic situation the only grid cell with wet deposition and also the grid cell is the only grid cell through which particles can go. First the physical process in nature is explained briefly and thereafter wet deposition in the backward in time simulations is considered.

Physically in nature the blue particles in Fig. 2a. are removed by wet deposition, green particles move with the wind above the cloud and red particles appear from another location after the cloud and rain. In the simulation the blue particles do not exist and only the red and green particles are released at the beginning of the backward in time simulation. The release time and the beginning of the simulation is marked by X in Fig 2b. In the simulation, the green particles go above the cloud layer and are not affected by wet deposition, while the red particles encounter the cloud and rain, and are therefore removed by wet deposition. At the start of the simulation the number of particles is $N_{red} + N_{green} = 11 + 3 = 14$. Due to wet deposition, the number of particles must decrease as the simulation time increases, but backwards in physical time (see arrows in Fig. 2b). Red particles are removed by wet deposition and by then the number of removed particles is $N_{red} = 11$. The red particles are in the grid cell at the beginning of the simulation, but not at the end of the simulation as wet deposition has acted to remove them. In practice this means that the particles "appear" or originate from the grid cell. To define the origin areas of the particles we want to know what percentage of particles are removed by wet deposition in the grid cell during the simulation and this is equivalent to knowing what percentage of particles originate in this grid cell in the real world. The percentage of particles originating from the grid cell is calculated by

$$\frac{N_{particles\ removed}}{N_{particles\ at\ the\ start}} = \frac{N_{red}}{N_{red} + N_{green}} = \frac{11}{11+3} = 0,79. \qquad (2.8)$$

Hence 0,79 originate from the grid in cell in this schematic situation.

2.1.3 FLEXPART's clustering process and plume trajectory calculations

The number of particles in FLEXPART simulations is usually tens or even hundreds of thousands. With this large amount of particles and multiple time steps, the quantity of output data is large. Stohl et al. (2002) presented cluster analysis, introduced by Dorling et al. (1992), which is a comprehensive statistical technique to condense FLEXPART output data and represent the FLEXPART output as trajectories. The basic idea behind the cluster analysis is to cluster particles to a number of groups at every output time step, and only write out the clustered particle positions instead of positions of each particle. These clustered particle positions form plume trajectories, which describe the paths of clustered particles during the simulation. The structure of the clustering process is as follows. First a seed for each cluster has to be generated. Seeds are positions that are generated every time step so that they cover the whole spread of particles in the time step and they describe the possible positions of the clusters. Secondly each particle at the time step is sorted to the seed which is the nearest one for it. Lastly the clusters' centroids, i.e. the center of masses, are calculated, based on the positions of the particles belonging to each cluster. These centroid positions are used then as the positions of the clusters.

The number of clusters is one of the parameters that the user can define in FLEXPART. The default value is five and it's also the value used in simulations presented in this thesis. Five clusters are chosen because Dorling et al. (1992) demonstrated that five clusters is the best option when the number of clusters is between one and seven. Eight or more clusters would be the best, but when handling less than 100 000 particles there wouldn't be enough particles per cluster if the number of clusters was eight or higher. Dorling et al. (1992) shows that when the number of clusters is five there is a local minimum in the Root Mean Square Deviation (RSMD) and after seven clusters the RSMD decreases again. RSMD describes the differences between predicted values and observed values and the smaller it is, the better the predicted values are.

The plume trajectory of an individual cluster, if considered independently of the other four, is not particularly useful. This is because the clustering process is done again at every output step. Since there is nothing that attaches individual particles to a specific cluster at each time step, particles can change the cluster, which they belong to, between two consecutive time steps. This means that between time steps, the fraction of particles belonging to each cluster can vary. To reduce this problem, the theory of mean trajectory is presented. The mean trajectory takes into account that the fraction of particles in each cluster is different in each time step and the mean trajectory is defined as the center of the mass of the clusters through the simulation. The difference between the mean trajectory and traditional backward trajectories is that the mean trajectory takes account of physical processes, for example turbulence and convection (Stohl et al., 2005).

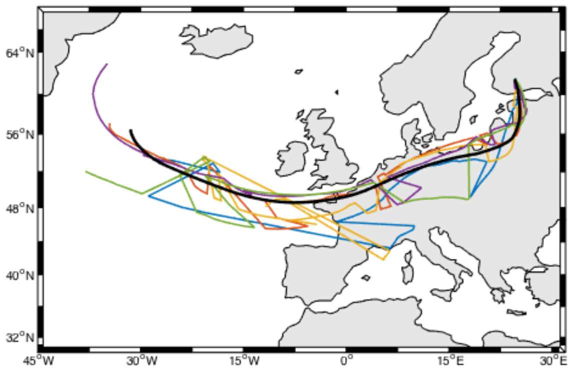

Figure 3: *Plume trajectories and mean trajectory from 96 hours backward in time FLEXPART simulation started at 08.02.2014 02UTC. Colored lines are plume trajectories and black line is the mean trajectory.*

In Figure 3 the trajectories and mean trajectory from one of the cloud cases (case 08.02.2014) are shown. Figure 3 shows that the paths of plume trajectories sometimes show non-smooth behavior i.e. their paths seesaw. This seesawing is caused by particles changing the cluster they belong to between time steps. When particle changes cluster between time steps it influences also the total mass related to each cluster. These differences between the mass related to each cluster at the time steps can not be noticed in the mean trajectory, because the calculation of the mean trajectory takes into account the mass of the different clusters. Figure 3 shows only the horizontal paths of the trajectories, but the clustering process is also done in vertical direction.

2.2 Hysplit introduction

Hysplit is a trajectory model developed by the National Oceanic and Atmospheric Administration (NOAA) Air Research Laboratory. The model has been originally developed to study atmospheric emergencies, case studies and climatology. The first version of Hysplit was released in 1982 (Draxler and Hess, 1998) and the model has been under development since and the present version is version 4. The fourth version of Hysplit can be run interactively in the internet or it can be downloaded to user's computer and run by using Graphical User Interface (GUI). Today's version has two applications: trajectories and forecast air concentrations (Draxler, 2014).

Hysplit is a hybrid model hybrid model between Eulerian and Lagrangian methods (Draxler and Hess, 1998). The trajectories and dispersion calculations in the model are Lagrangian, thus they follow along with a particle or puff, while the forecast concentration are Eulerian, thus they are calculated on fixed grid points. In here only the Lagrangian part of the model is considered because trajectories of Hysplit are under research.

The three-dimensional velocity field is the only meteorological input field required for trajectory calculations in HYSPLIT. The reason for this is that trajectories are calculated straight from advection by time integration. Hysplit can use as input data in many different vertical coordinate systems: pressure-sigma, absolute pressure, terrain-sigma or hybrid absolute-pressure-sigma coordinate system, and therefore it is not very selective for its meteorological input data. Usually these meteorological input fields used in Hysplit simulations have to go through some pre-processing because of the Hysplit's coordinate system is terrain-following. The horizontal output grid of Hysplit is always the same as the grid of the meteorological input. Hysplit supports three conformal map projections: Polar Stereographic, Mercator and Lambert Conformal. The results of Hysplit have been evaluated in many different studies. For example, trajectories have been evaluated against Aerosol Characterization Experiment (ACE) balloon trajectories where the trajectories of the model were compared with the measured path of three balloons (Draxler and Hess, 1998). The results from the evaluations have been good and consistent.

NCEP's Data Assimilation System (GDAS) meteorological data has been used as meteorological input data for Hysplit in the simulations presented in this thesis. Time resolution of GDAS is 3 hours, horizontal resolution is one degree by one degree and the model has 23 vertical layers. GDAS data is gridded observation data from, for example, surface observations, aircraft reports and satellite observations which goes throw data assimilation processes.

Hysplit outputs are the coordinates of ensemble trajectories. In Hysplit the ensemble trajectories are produced by differing the initial position by one grid point in meteorological grid in horizontal way to each possible direction and 0,01 sigma units in the vertical way up and down, 0,01 sigma units is about 250 meters. By differing the initial position 27 ensemble trajectories are produced to describe the motions of particles.

3 Case studies and diagnostic

3.1 Case studies: overview and justifications for selection

Seven interesting cloud situations and seven new particle formation situations used as case studies were selected based on the interesting observations made in The Biogenic Aerosol – Effect on Clouds and Climate (BAECC) campaign. BAECC was 8-month long intensive measurement campaign in SMEAR II (Hyytiälä, Finland) from February 2014 to Semptember 2014. The main aim of the campaign was to understand what is the role of biogenic aerosols in cloud formation (Petäjä et al., 2016). Based on Mason (1971) the majority precipitation in Finland is initiated by ice-phase processes and to study the surface observations of solid precipitation the campaign included BAECC Snowfall Experiment (SNEX) campaign which concentrated on snowfalls.

Cloud cases are from times when interesting cloud events were observed and aerosol cases are from the mornings when interesting or sporadic new particle formation was observed. All of the case studies are from the time period from February 2014 to April 2014. Specific dates and times of case studies are listed in Table 1. Dates and times in Table 1 are the times when 96 hours backward in time simulation were started. Later on in this thesis the simulations are referred only by the date on which the simulation was started.

Table 1: *Dates and times of case studies used in this thesis. Case studies bases on the observations made in BAECC and BAECC-SNEX campaigns.*

cloud cases	simulation started	aerosol cases	simulation started
02 February	17:00 UTC	08 April	08:00 UTC
08 February	02:00 UTC	22 April	08:00 UTC
16 February	01:00 UTC	23 April	08:00 UTC
22 February	00:00 UTC	24 April	08:00 UTC
03 March	08:00 UTC	25 April	08:00 UTC
08 March	05:00 UTC	26 April	08:00 UTC
18 March	12:00 UTC	27 April	08:00 UTC

There were a lot of differences in the characteristics of the synoptic situations between the cloud and aerosol cases. The main difference was that during cloud cases there was a low pressure system or systems over Northern Europe influencing the flow field and during aerosol cases there was a high pressure system or systems over Northern countries.

3.2 FLEXPART

3.2.1 Set-up control and sensitivity experiments

All of the case study simulations were made three times with FLEXPART, with three different tracer particles, the justification for the different simulations and particles is presented in paragraph below. Below there are also briefly described the main features of the options used in the FLEXPART simulations and the Appendix B contains the used option files.

First the simulations were made with passive tracer particles. Passive tracer particles passively move with the wind and removal processes do not affect passive tracer particles. The passive tracer particle simulations were conducted for the model comparison and passive tracer particles was used to made the FLEXPART simulations as close to the Hysplit simulations as possible. Secondly the simulations were made with default wet deposition tracer particles (WDT1). WDT1 particles are used in simulations to identify areas where wet deposition occur. Identifying is possible because WDT1 particles are strongly affected by the wet deposition parameterization, both in-cloud and below-cloud scavenging are influencing. Third simulations were also made with wet deposition tracer particles but the below-cloud scavenging parameterization of the particles was set to as small as possible by setting scavenging coefficient A and below-scavenging parameter B in Equation 2.1 such that the result of the equation comes close to zero. These particles with small below-cloud scavenging parameterization are called WDT2 particles. When the the result of Eq. 2.1 comes close to zero below-cloud scavenging can be simplified to be switched off because it is so small. The values of scavenging coefficient A and below-scavenging parameter B used for each particle are listed in Table 2. Simulations with WDT1 and WDT2 particles were conducted to study the second aim of thesis, the effect of wet deposition on the mass transported to Finland. Simulations conducted with WDT1 focused on the general characteristics of wet deposition and simulations conducted with WDT2 concentrated the effect of small below-cloud scavenging parameterization.

Table 2: *Scavenging coefficients (A) and scavenging parameters (B) used to calculate the below-cloud scavenging parameter for different tracer particles (Equation 2.5). Negative value of scavenging coefficient A for passive tracer in is the way how wet deposition parameterization is switched off in FLEXPART model.*

Tracer	A	B
Passive Tracer	-9.9^{-09}	-
Wet Deposition Tracer I	$1 * 10^{-04}$	0,62
Wet Deposition Tracer II	$0,1 * 10^{-40}$	0,00

In all of the simulations made with FLEXPART the particles were released instantly. At the instant release all of the particles are released exactly at the same time. These instant release times are based on the observation times and the times are listed in Table 1. The number of particles released in each FLEXPART simulation was 50 000. In the cloud cases the release layer was from 0 to 500 meters and in aerosol cases the release altitude was 100 meters. The release heights of cloud and aerosol cases are different to each other because usually the cloud depth is higher than 100 meters and the particles wanted to be at least partly released in-cloud during the cloud cases. The particles wanted to be released in-cloud was that in-cloud they could represent cloud droplets and to fulfill this assumption the release altitude had to be higher than the cloud base. For aerosols cases the release altitude of 100 meters was optimal. The release area in FLEXPART simulations was 0,02 times 0,02 degrees wide, from the latitude 61.840°N to the latitude 61.860°N and from longitude 24.270°E to longitude 24.290°E (A = 2,35 km^2). Subgrid scale parameterizations and convection parameterization were set to be on during the FLEXPART simulations. The output time interval of the FLEXPART simulations was one hour and the output grid was 0,2 times 0,2 degrees large over the whole world.

The FLEXPART outputs used in this thesis are trajectory output and particle information output. From the trajectory output files the all information required to calculations described below was only the latitudes and longitudes of the plume trajectories at each time step. FLEXPART particle information output includes details from all of the particles and it is an optional output in the same way as the trajectory output. In the case study simulations the particle information output was saved after each time step so that the differences between time steps could be studied, the mass and altitude of each particle at each time step were needed to calculate the total scavenged mass in the simulations and the removal altitudes.

3.2.2 Diagnostics

In this section diagnostics from FLEXPART will be described. First a method to handle the spread of plume trajectories is described. Lastly diagnostics concerning particle mass and altitude are presented.

The spread of the plume trajectories at each time step can be calculated and used to estimate the variability of the source areas of the particles. If the particles are from a larger area the plume trajectories are widely scattered, while in the case where particles are from a smaller area the plume trajectories are narrowly scattered. There are many statistical tools that can be expanded to represent this spread of the plume trajectories. For instance, by calculating standard deviations of the latitudes and longitudes of the plume trajectories at each time step, the spread could be defined in the vertical and horizontal directions. In the case where the spread of plume trajectories is defined in two directions, to analyze the differences between the cases both directions have to be regarded. When the number of cases increases, comparing of the spread estimates becomes more difficult if there are two variables that together represent the total spread. To avoid the difficulty with two variables, the spread was wanted to display with only one variable which incorporates both north-south and west-east dispersion. This variable was chosen to be a minimum radius of the circle that encloses the plume trajectory locations at each time step. The minimum radius of the circle is calculated in kilometers to describe the area of the circle where plume trajectories are spread.

When knowing the mass of each particle at each time step, the change of the mass of the particle caused by scavenging at a time step can be calculated by

$$\Delta m = m(t) - m(t + \Delta t) \tag{3.1}$$

where Δm is the scavenged mass, $m(t)$ is the mass before the time step Δt and $m(t + \Delta t)$ is the mass after the time step Δt. By integrating twice, first over all of the particles and second over all of the time steps the total scavenged mass during the simulation can be defined.

To define the removal altitudes of the particles the troposphere between the ground and 5 kilometers was sliced into 50 meter thick layers. After each time step Δt the altitude of each particle was read and it was checked to which of these 50 meter layers the particle belonged. When the right layer where the particle belonged to was found, the scavenged mass of the particle was added to the total sum of scavenged mass in that layer. This process was done for all of the particles after all of the time steps. The result of this process is the total scavenged mass as a function of altitude integrated over the whole simulation.

In backward in time simulations the concept of the mass of the particle is not quite clear and the scavenged mass does not have a physical meaning. For this reason the total scavenged mass during the simulation and the scavenged mass as a function of altitude are divided by the release mass, i. e. the total mass of the particles at the release time. In this way the results are dimensionless when the dimension is kg/kg.

3.3 Hysplit

3.3.1 Hysplit experiments

All of the case studies were simulated once with Hysplit and 96 hours backward in time simulations were started at the given times listed in Table 1. The release height in each Hysplit simulation was 100 meters. The output of Hysplit is the coordinates of ensemble trajectories. Below the mean ensemble trajectory calculation method is described.

3.3.2 Diagnostics

Ensemble means are calculated from ensemble trajectory coordinates by the process described below. Ensemble means were calculated in polar coordinates because in polar coordinates the spherical shape of Earth can be ignored. In polar coordinates each coordinate point on Earth can be presented by function of the azimuth angle and range. For the ensemble means the arithmetic means of the azimuth and range were calculated separately at each time step. After the means were calculated in polar coordinates they could be converted to the latitude and longitude coordinates and the ensemble means were calculated. When converting trajectories to and from polar coordinates, SMEAR II (Hyytiälä, Finland) location was used as the center point. In addition to the geographical coordinates of the ensemble trajectories and ensemble mean trajectories the data given for analyses included also the altitudes of the ensemble mean trajectories at each time step.

Spread of the Hysplit ensemble trajectories is determined in the same way as the spread of FLEXPART plume trajectories: The radius of the minimum circle that encloses the trajectory locations at the time step. The calculation method of the radius of minimum circle was exactly the same as the calculation method of the radius of minimum enclosing circle of FLEXPART plume trajectories.

4 Results from trajectory studies

The results of this chapter are related to the first aim of the thesis. The aim is to investigate how FLEXPART can represent the source areas of particles and study how source areas, represented in FLEXPART and Hysplit, differ from each other. The aim is approached first by studying the spread of FLEXPART plume trajectories. Spread describes how well the mean trajectory represents the source area of particles in the simulation. If the particles are from a narrow source area, spread is smaller and the mean trajectory represents all particles well. In situations where the particles are from a large source area and the spread is wider the mean trajectory does not necessarily represent the source area of all particles very well. Spread naturally varies between cases, for example, because the synoptic situation can be different between the cases and thereby the wind directions and strength are different. To represent the magnitude of the spread during this chapter the minimum radius enclosing circle variable is used. The variable is presented and described in Chapter 3.2.2. Secondly in this Chapter the spread differences of FLEXPART plume trajectories and Hysplit ensemble trajectories are discussed before lastly the mean trajectories of FLEXPART and Hysplit are studied by comparing the altitude and geographical location of mean trajectories. All topics are discussed by analyzing the cloud cases and aerosol cases separately and after that by comparing the general characteristics of cloud cases to the general characteristics of aerosol cases.

FLEXPART simulations analyzed in this section were conducted using passive tracer particles so that the results can be fairly compared to Hysplit simulations. This is because in the Hysplit simulations there was no wet deposition affecting the particles.

4.1 Spread of trajectories

4.1.1 Spread of FLEXPART plume trajectories

Figure 5 shows the radii of minimum enclosing circles from the FLEXPART cloud case simulations at a function of time. In Figure 5 there can be seen randomly looking, usually downward peaks, in the calculated radii. For instance, in the 18.3.2014 and 8.2.2014 cases there are several peaks between 42 and 96 hours of the simulation. The peaks are caused by the behavior of the clustering process in specific types of synoptic situations. By comparing the synoptic charts to the location of clusters at the time steps when the peaks were observed three synoptic factors were identified, that might be separately or together reasons for this kind of noisy behavior where peaks appear. All of these three factors are synoptic phenomena that spreads particles to a wide area and the clustering process responds to the spread of particles. The response of clustering process is as follows. The clustering processes tries to make the cluster so that the clusters cover the whole area where particles are detected but at the same time, the clustering processes do not depend on the cluster's position at the previous time step. When the clustering process tries to cover the whole area where particles are spread the positions of the clusters' seeds are generated to back and forth positions between successive time steps while the particles spreads to the wider area.

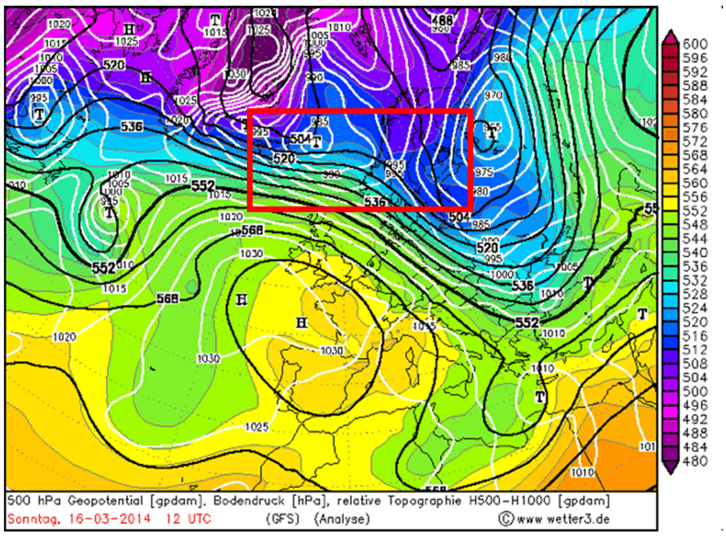

Figure 4: *The Global Forecast System (GFS) analyses valid at 16.3.2014 12UTC. Black contours are 500 hPa geopotential height, white contours are surface level pressure and colours describe the thickness between 500 hPa and 1000 hPa pressure surfaces. The red box shows the area where positions of clusters were observed between 45-51 hours of the 18.3.2014 simulation.*

All of three factors that are thought to be the reasons for the peaks observed in the spreads of cloud case simulations were observed in synoptic maps at 16.03.2014 12 UTC, which is 48 hours into the backward simulation which started at 18.3.2014. Figure 4 shows the synoptic situation at 16.03.2014 12 UTC based on The Global Forecast

System (GFS) analyses. Based on Figure 4 the following three factors causing noisy behavior can be identified:

1. Large angle between surface level pressure isobars and 500 hPa geopotential height contours. The reason for the noisy behavior in these situations may be caused by particles with different altitudes. The wind direction at 500 hPa is different compared to the wind direction near the surface, also the wind speed is usually higher in the upper atmosphere (Lackmann, 2012) so the particles in the upper level travel longer distances than the particles near to the surface in a same time. In the red box area of Figure 4 this factor can be detected near the Swedish-Norwegian border.

2. Very small gradients in either, or both, the surface level pressure isobars or 500 hPa geopotential height contours. In the areas of small gradients wind directions can vary a lot in a small area. The wind speeds are in area of small gradient are however weak. Despite the weak wind speeds the varying wind directions in small gradient areas spreads particles to the wide area in for example a couple of days. In the red square area of Figure 4 this can be seen over Gulf of Bothnia.

3. There are two low or high pressure systems close to each other and the particles are influenced by both of them. In Figure 4, this situation can be seen. There are two surface low pressure systems, one over Western Russia and one over the Norwegian sea. The 995 hPa isobars of both systems passes very close to each other in the red square marked in Figure 4. In this region, a small change in a particles position potentially results in a very large change at a later time.

The relative importance of these three processes can not be determined neither the impact of these three factors on the radii can be quantified.

In general the spread of plume trajectories is wide in each cloud case but there is a large amount of variation between the cases: the difference between the smallest and the largest radius during the last three time steps is about 1 450 kilometers (Fig. 5). After 48 hours the difference between the smallest and the largest radius is 550 kilometers. Thus the difference in the size of calculated radii increased more during the last 48 time steps than during the first 48 hours of

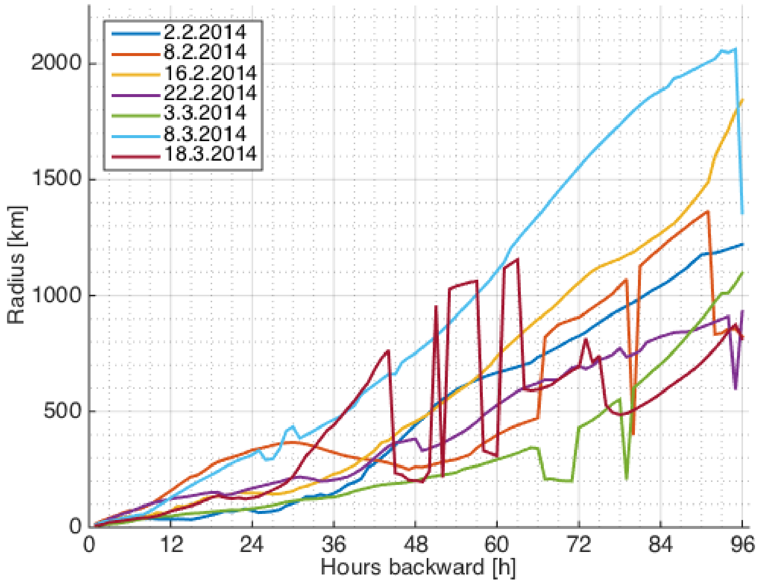

Figure 5: *Spread of plume trajectories as a function of time in FLEXPART cloud case simulations demonstrated with radius of minimum enclosing circle.*

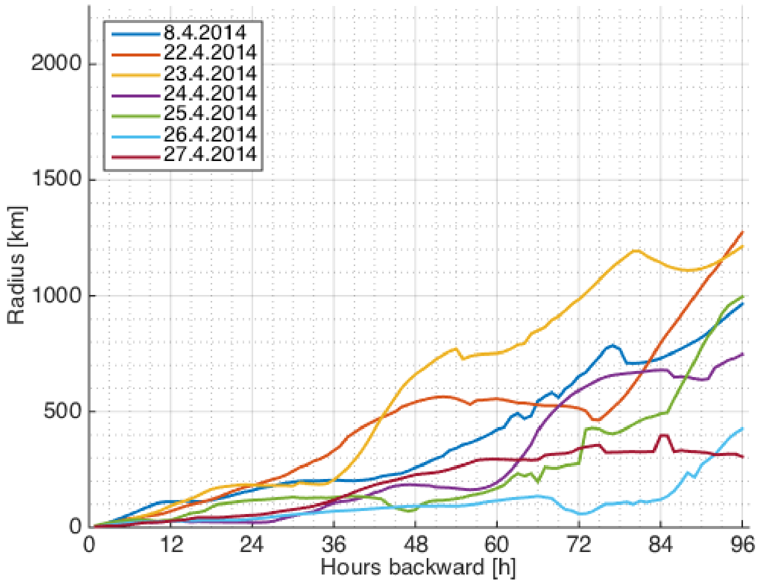

Figure 6: *Spread of plume trajectories as a function of time in FLEXPART aerosol case simulations demonstrated with radius of minimum enclosing circle.*

the FLEXPART cloud case simulations. In other words during the first half of the simulations the calculated radii of all of the simulations are quite similar but during the second half of the simulation the calculated radii differ more from each other. When the sizes of the radii vary this much between individual cases, it is not possible to make an exact definition to describe the spread of cloud cases after the 96 hour simulations. A loose definition could be that by the end of the 96 hour simulation the radius size in cloud cases is over 1 000 kilometers or even larger.

Figure 6 shows radii of minimum enclosing circles from all of the aerosol cases as a function of time. Figure 6 shows that in aerosol cases there is little noisy behavior in the sizes of radii despite that during all of the aerosol cases there was high pressure and anticyclone conditions over the Northern countries. A small pressure gradient near the high pressure center is a common feature related to high pressure systems and this feature was observed during a few of the simulations but strong noisy behavior were still not detected. Based on this it can be assumed that the effect of this factor (weak pressure gradient, number 2. in the list) is weaker than the effect of numbers 1. and 3. in the list. It can be also just a coincidence that the weak pressure gradient is detected during the noisy behavior in cloud cases and by that named as a factor causing the noisy behavior.

In aerosol cases the variations in the sizes of radii between cases is smaller than in the cloud cases: the difference between the smallest and largest radii during the last three hours in aerosol cases is about 1000 kilometers. After 48 hours the difference is 550 kilometers which is the same as in the cloud cases. The difference between cases then increases slightly more during the first 48 hours of the simulations than during the last 48 hours of the simulations. The spread in aerosol cases increases quite constantly based on these simulations. For aerosol cases the spread after the simulations could be described as that the sizes of radii are after the 96 hours simulations under 1500 kilometers.

In general, the spread after a specific amount of hours is higher in cloud cases than in aerosol cases. Also the variation inside the cases is larger in the cloud cases than in aerosol cases. The spread increases more evenly over the whole simulation in aerosol cases, when in cloud cases increasing is stronger in the last half of the simulation. The first 24 hours of the spreads are quite similar between the cloud and aerosol cases but after that the differences start to grow. That the spread increases much more in the last 48 hours of the cloud cases than in the aerosol cases shows that in cloud cases the particles are from larger source area than in aerosol cases and in cloud cases there are some synoptic features that gather the particles together when they are coming forward in time to the SMEAR II (Hyytiälä, Finland).

Because in aerosol cases the particles are from more narrow an area the particles have to go with the flow in a body instead of some synoptic situation gathering them to SMEAR II (Hyytiälä, Finland) forward in time.

4.1.2 Comparison between FLEXPART and Hysplit

Figure 7 shows the differences in the sizes of calculated radii between the cloud case simulations of FLEXPART and Hysplit. When the difference is positive, the spread of trajectories at the time step is larger in FLEXPART than in Hysplit, whereas when the difference is negative the spread of trajectories at the time step is larger in Hysplit than in FLEXPART. Increasing difference with time refers to the situation where the spread increases more rapidly in FLEXPART than in Hysplit. When the difference decreases with time the spread of Hysplit increases more than the spread of FLEXPART. If the difference is constant it refers to the situation where the spread of the trajectories increases with time equally fast in both models.

The noisy behavior in Figure 7 is the same noisy behavior that is visible in Figure 5. Hysplit does not cause noisy behavior because the ensemble process is different to the clustering process as previously noted in Chapters 2.1.3 and 2.2. Figure 7 shows that during the first 48 hours of simulations the spread was generally larger in Hysplit during the cloud cases. After 48 hours the cases where there was no noisy behavior (2.2.2014, 16.2.2014 and 8.3.2014) the spread of FLEXPART increases and be comes larger than the spread of Hysplit. At the same time, those cases (8.2.2014, 3.3.2014 and 8.3.2014) where there was a lot of noisy behavior, the spread is much larger in Hysplit than in FLEXPART during the last 48 hours. The 22.2.2014 case is the only case where there is not a large difference between the spreads of Hysplit and FLEXPART. The maximum difference in the spreads is in the 22.2.2014 case about 150 kilometers, which is less when comparing to other cloud cases.

Figure 8 shows the differences in the size of radii between the aerosol case simulations of FLEXPART and Hysplit. Based on Figure 8 the spread in aerosol cases is generally higher in Hysplit than in FLEXPART. In a few of the cases at the beginning of the simulations the spread of FLEXPART trajectories is slightly larger than the spread of Hysplit trajectories and in one case (23.4.2014) the differ-

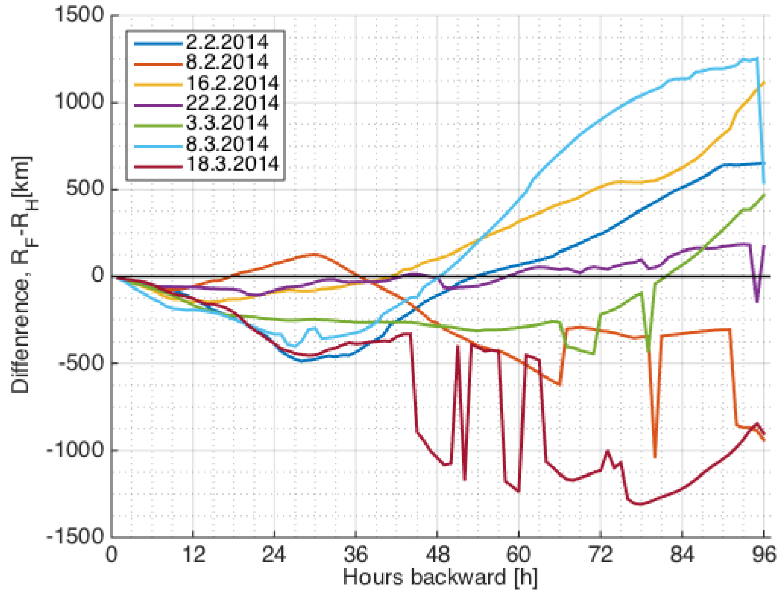

Figure 7: *Differences between radius of minimun enclosing circle of FLEXPART plume trajectoris and radius of minimun enclosing circle of Hysplit ensemble trajectories as a function of time in cloud case simulations.*

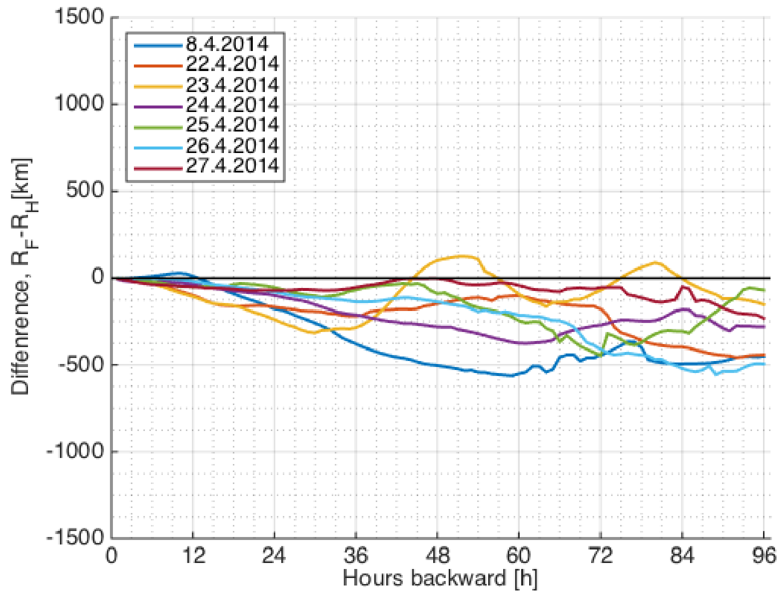

Figure 8: *Differences between radius of minimun enclosing circle of FLEXPART plume trajectoris and radius of minimun enclosing circle of Hysplit ensemble trajectories as a function of time in aerosol case simulations.*

ence increases above zero twice but both times the difference in radii is less than or equal to 100 kilometers. The maximum difference between the radii of the trajectories is about 550 kilometers in aerosol case simulations.

Generally during both cloud and aerosol cases the spread is larger in Hysplit. In aerosol cases the difference between FLEXPART and Hysplit is smaller than in cloud cases.

4.2 Mean trajectories

4.2.1 Differences in the altitudes of mean trajectories

In addition to the longitudes and latitudes of plume and ensemble trajectories both models give as an output variable which describes the altitudes of the trajectories. In FLEXPART also the altitude of the mean trajectory is given as an output but in Hysplit the altitude of the mean trajectory is calculated as described in Chapter 3.3.2. The altitude is an important factor for the particles motions because the wind speed usually varies between the altitudes (Lackmann, 2012) which can seen from the soundings (Fig. 9 and 10). The humidity and wind profile soundings (Fig. 9 and 10) were made in SMEAR II (Hyytiälä, Finland) during BAECC and BAECC-SNEX campaigns. The times of the used soundings are chosen by taking the soundings that are the closest to the starting times of the simulations. The vertical soundings are considered here because they describe the vertical structure of the atmosphere. Vertical structures of wind and humidity are important because based on them many assumptions related for example, to clouds and trajectories in different altitudes can be made.

Figure 11 shows that in cloud cases the mean altitudes of the FLEXPART and Hysplit mean trajectories are at totally different altitudes, the altitudes are not in any stage within the error bars. There is also only a little overlap in the range of heights between FLEXPART and Hysplit near the 54-84 hours of the simulation. The mean and the error bars of the mean are in FLEXPART closer to the maximum altitude than minimum altitude, while in Hysplit the mean and the error bars are almost at the middle of the maximum and minimum altitudes. This refers to there being more trajectories of cloud cases closer to the maximum altitude than minimum altitude in FLEXPART while in Hysplit the altitudes of mean trajectories are evenly distributed between minimum and maximum altitudes in cloud cases. In conclusion all of these factors presented above indicate that the mean trajectories of FLEXPART are many hundreds of meters higher in altitude than

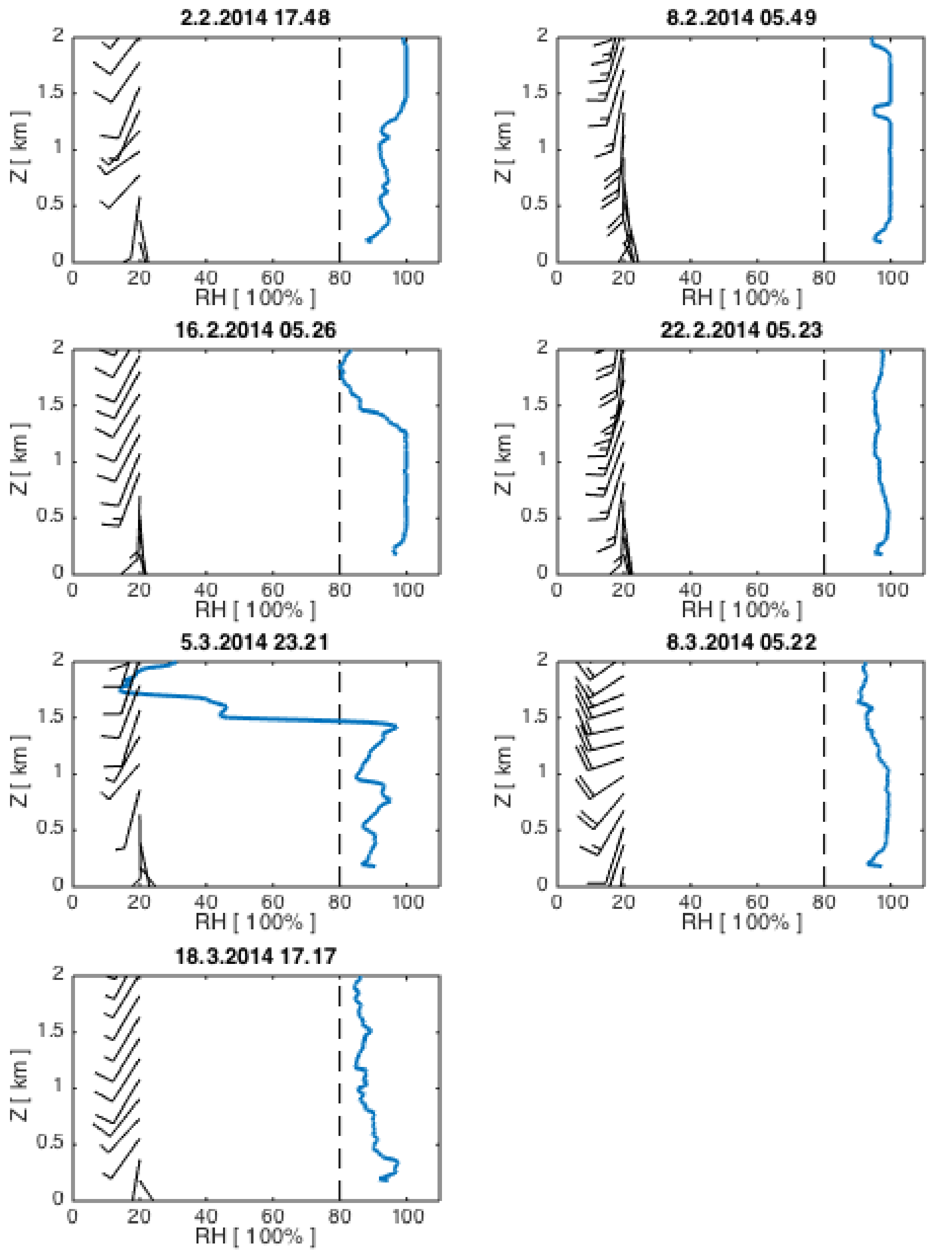

Figure 9: *Wind and humidity soundings made during BAECC and BAEEC-SNEX campaigns. The soundings presented in here are chosen so that their starting times are close to the starting times of the cloud case simulations. The dotted line represents 80 % humidity. Wind directions and wind speeds are presented by Wind Barbs, small barb indicates the wind speed less than 5 m/s, long barb indicates wind speed* $5\ m/s \leq U < 10\ m/s$*, long barb and small barb indicates wind speed* $10\ m/s \leq U < 15\ m/s$ *and so on.*

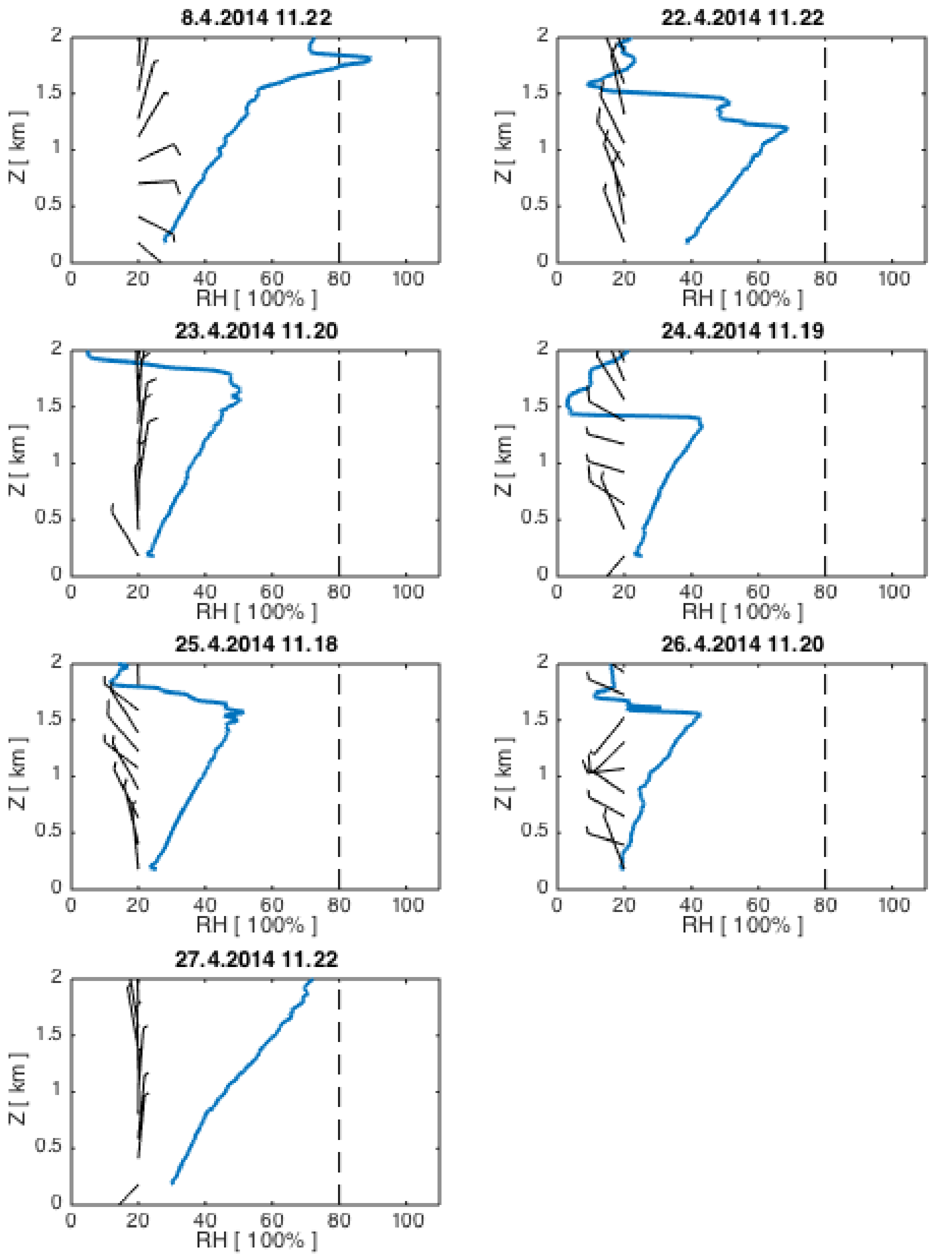

__Figure 10:__ Wind and humidity soundings made during BAECC and BAEEC-SNEX campaigns. The soundings presented in here are chosen so that their starting times are close to the starting times of the aerosol case simulations. The dotted line represents 80 % humidity. Wind directions and wind speeds are presented by Wind Barbs, small barb indicates the wind speed less than 5 m/s, long barb indicates wind speed $5\ m/s \leq U < 10\ m/s$*, long barb and small barb indicates wind speed* $10\ m/s \leq U < 15\ m/s$ *and so on.*

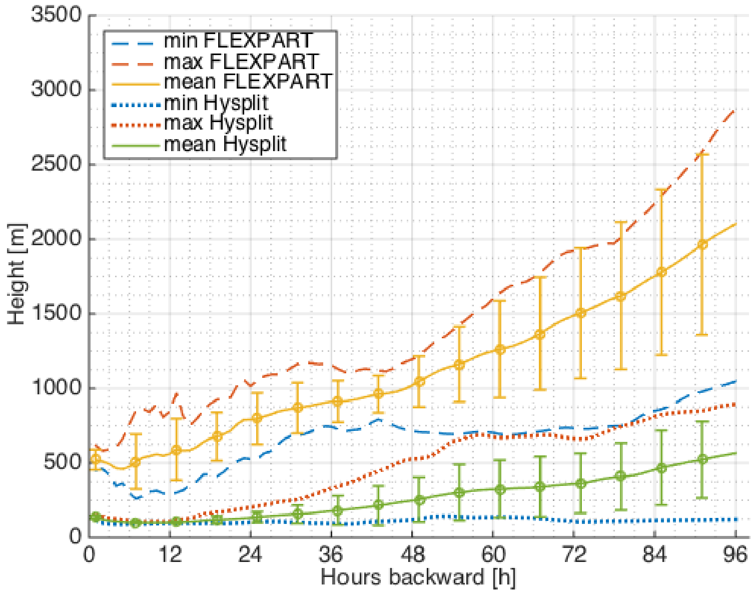

Figure 11: *Mean, minimum and maximum altitudes of FLEXPART and Hysplit mean trajectories in cloud cases as a function of time. Mean is an arithmetic mean of the altitudes of the seven mean trajectories and the error bars for the mean are the standard deviation of the seven altitudes. The minimum is the minimum altitude of the mean trajectories after each hour, so the minimum does not describe one specific trajectory and the trajectory of which the minimum altitude is from can vary between the hours. The maximum is the same than the minimum, but with maximum values of the mean trajectory altitudes. The release height was in FLEXPART 0-500 meters and in Hysplit 100 meters.*

the mean trajectories of Hysplit during the cloud cases. Also based on Figure 11 in FLEXPART there is more variation in the altitudes between the cases, and also the difference between the minimum and maximum altitudes at the end of simulations is more than two times larger in FLEXPART than in Hysplit.

Figure 12 shows that in aerosol cases the minimum and maximum altitudes of the mean trajectories have smaller differences between the models than in cloud cases. In the maximum altitudes there are more difference between the models and the difference between the maximum altitudes varies a lot during the 96 hours. The means of FLEXPART and Hysplit mean trajectory altitudes are an almost equal distance from each other during the aerosol simulation which means that the mean altitudes increase similarly. Also the means are, despite the first 12 hours, within the error bars which indicates that in some cases the trajectories of FLEXPART and Hysplit might be at the same altitude. In FLEXPART the mean altitude and error bars are a

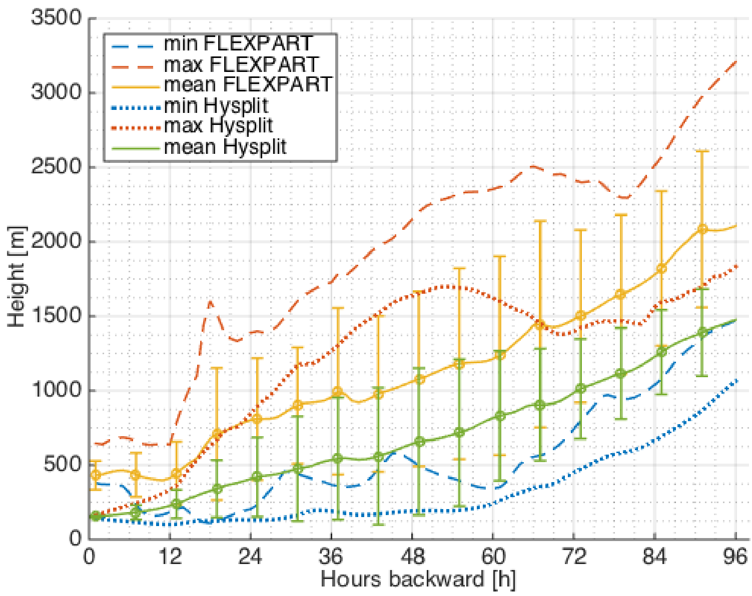

Figure 12: *Mean, minimum and maximum altitudes of FLEXPART and Hysplit mean trajectories in aerosol cases as a function of time. Mean is an arithmetic mean of the altitudes of the seven mean trajectories and the error bars for the mean are the standard deviation of the seven altitudes. The minimum is the minimum altitude of the mean trajectories after each hour, so the minimum does not describe one specific trajectory and the trajectory of which the minimum altitude is from can vary between the hours. The maximum is the same than the minimum, but with maximum values of the mean trajectory altitudes. The release height was in both models 100 meters.*

little bit closer to the minimum altitude than the maximum altitude, which indicates that the are more FLEXPART mean trajectories close to the minimum altitude than the maximum altitude. The variation between the aerosol cases is larger in FLEXPART, after 96 hours; the difference between maximum and minimum altitudes at the end of the simulations is about twice as large in FLEXPART compared to the difference in Hysplit and the error bars of FLEXPART are at least twice as large compared to the error bars of Hysplit after 96 hours.

Both in cloud and aerosol cases the altitudes of mean trajectories are generally higher in FLEXPART than in Hysplit (Fig. 11 and 12). Also it is important to notice that the mean altitudes of FLEXPART are almost the same in aerosol and cloud cases. For instance, after 24 hours the mean altitude is in both, cloud and aerosol cases, about 750 meters, after 72 hours the mean altitude is 1 500 meters in both and at the end of the simulations the mean altitudes are about 2,1 kilometers. In Hysplit the mean altitudes of cloud and aerosol cases do not resemble each other, in aerosol cases the mean altitude is much higher than in cloud cases. Based on these the altitudes of mean trajectories vary more in the Hysplit simulations than in the FLEXPART simulations. In both models the variation inside the cloud cases is larger in aerosol cases, based on the sizes of the error bars.

The release heights were in Hysplit 100 meters in both cloud and aerosol cases. In FLEXPART the release height was 100 meters in aerosol cases and 0-500 meters in cloud cases. Given that when the release height is the same (aerosol cases) the mean altitude in Hysplit is similar to that in FLEXPART, but when the release height is different (cloud cases) a larger difference in mean altitudes exists, it can be suggested that the source height is important to the altitudes of the trajectories. If the humidity is constant within the height it can be said that the layer is mixed, based on the humidity soundings of cloud cases (Fig. 9) and aerosol cases (Fig. 10) the layer from ground to 2 kilometers is more mixed in cloud cases than in aerosol cases because in cloud cases the humidity is in most of the cases constant within the height, while in most of the aerosol cases there can be observed local maximums in humidity near 1,5 kilometers. In a well mixed layer the turbulence can spread the particles over the whole layer (Stuhl, 2015), which might have happened in FLEXPART cloud case simulations. Hysplit, and other traditional trajectory models, can not represent the turbulence as well as the Lagrangian particle dispersion models, which can be led to the higher trajectory altitudes in FLEXPART than in Hysplit during the cloud case situations.

The differences in the altitudes can also partly be the reason for the differences between the spreads of FLEXPART and Hysplit cloud case simulations discussed in Chapter 4.1.2. Based on the soundings (Fig. 9 and 10) the wind direction varies quite slowly within the height, but when the difference between altitudes is high the differences in wind directions can be significant. This difference in wind directions means that the particles which are at totally different altitudes can spread to totally different directions while the air parcels that are closer to each other can not spread to totally different directions.

4.2.2 Differences in geographic locations of mean trajectories

Figure 13 shows that there are significant differences between geographic locations of FLEXPART and Hysplit mean trajectories during the cloud cases. For example, in the 8.3.2014 case the difference between the endpoints of the mean trajectories was 5064 kilometers. During most of the cloud cases the directions of trajectories are the same, for instance in the 3.3.2014 case both trajectories are from South and the endpoints are over southern Europe and in the 18.3.2014 case trajectories of both models came from the Barents Sea and Kola Peninsula. In the 22.2.2014 case there is the largest difference between the direction of trajectories when the FLEXPART mean trajectory ends over Denmark and Hysplit mean trajectory ends over Southern Lapland. In generally the trajectories of Hysplit are shorter than the trajectories of FLEXPART. This is probably mostly a result of the different altitudes of the trajectories, as presented in Chapter 4.2.1. Figure 11 shows that in FLEXPART the mean altitude of mean trajectory altitudes is between 500 meters to 2,1 kilometers while in Hysplit the mean altitude of mean trajectory altitudes is between 100 meters to 550 meters. Figure 9 shows that the wind speeds in cloud cases are generally much higher over 500 meters than below it. For example, in the 2.2.2014 case the wind speed was less than 5 m s^{-1} below 500 meters and in altitude of 2 kilometers the wind speed was over 10 m s^{-1}. The difference of over 5 m s^{-1} in wind speeds is large, it means that the particle with higher wind speed can travel more than 18 kilometers further in one hour than the particle with lower wind speed. When there are 96 hours in the simulations the particle with higher wind speed can travel 1728 kilometers further than the particle with lower wind speed but with the same wind direction in the same simulation.

Figure 14 shows that during the aerosol cases there are only slight, maximum of hundreds of kilometers, differences between the mean trajectories of FLEXPART and Hysplit. The smaller differences in aerosol cases than in cloud cases may be explained with Figures 12 and 10. Figure 12 shows that the mean altitudes of the FLEXPART and Hysplit mean trajectory altitudes are mostly within the error lines during the simulations. This indicates that the trajectories of FLEXPART and Hysplit have been almost at the same altitudes. Also the

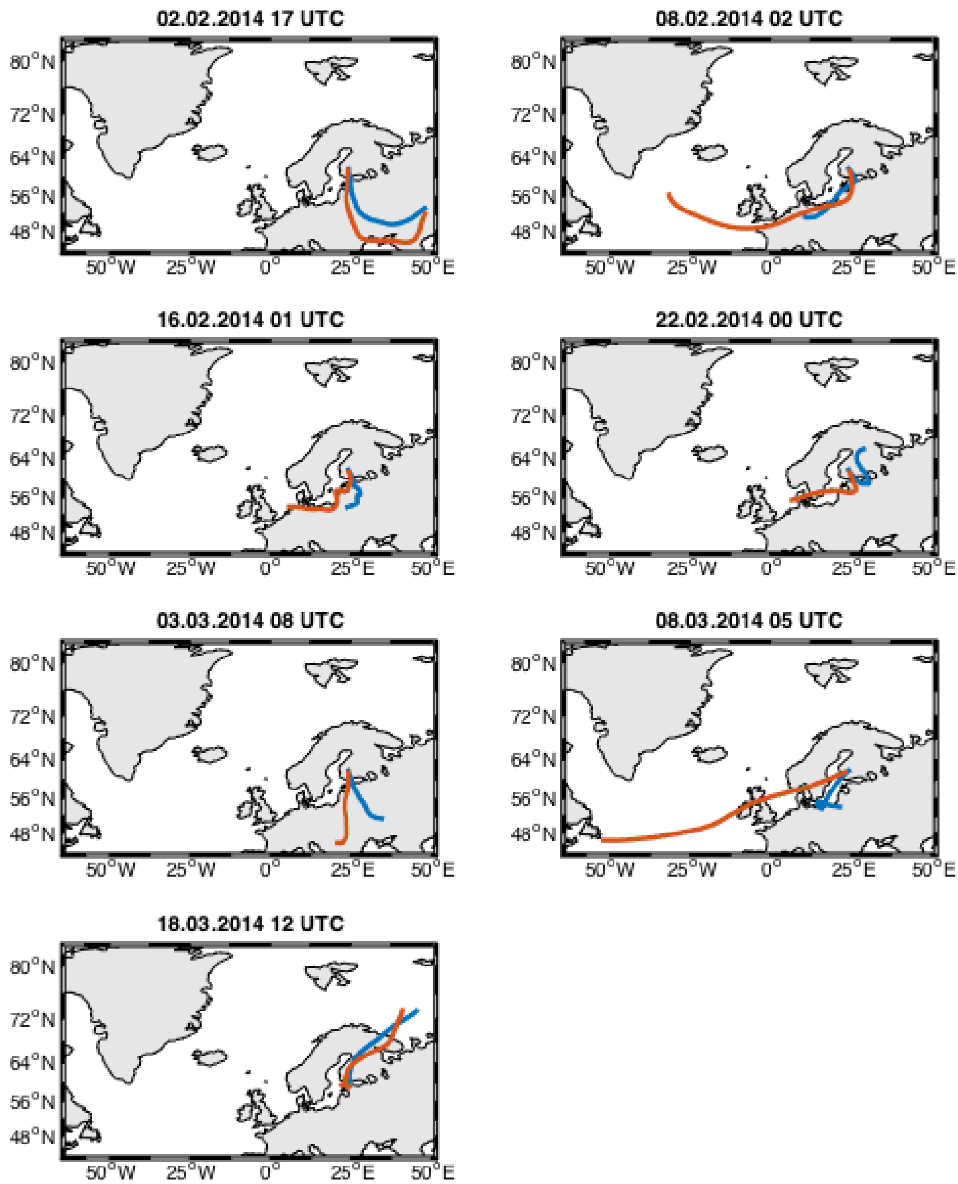

Figure 13: *96 hours FLEXPART mean trajectories (orange lines) and Hysplit mean trajectories (blue lines) from cloud case simulations.*

differences in the wind speeds in different altitudes are smaller in the aerosol cases than in the cloud cases based on Figure 10, which shows that the wind speeds are in aerosol cases in the layer from the ground to 2 kilometers mostly between 0-15 m/s. The smallest differences in the locations of mean trajectories occur in the 25.4.2014 case, where

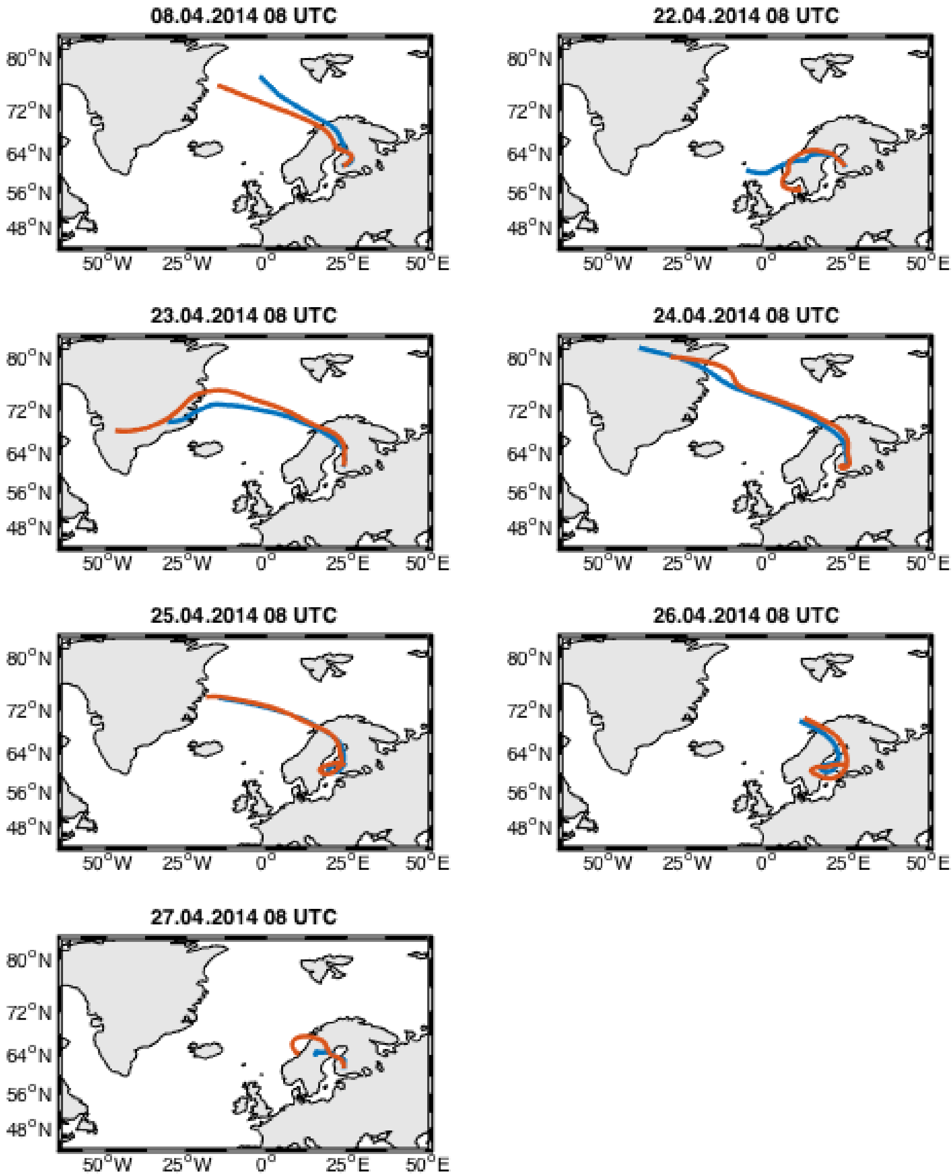

Figure 14: *96 hours FLEXPART mean trajectories (orange lines) and Hysplit mean trajectories (blue lines) from aerosol case simulations.*

the trajectories are exactly co-located, even though the trajectory of Hysplit ends before the trajectory of FLEXPART. In every aerosol case the biggest differences between the geographic locations of trajectories can be detected closer to the endpoints of trajectories than to the release point. The largest difference are closer to the endpoints than the release points because of, based on Stohl (1998), the errors in trajectories increases while the time increases backward. The increasing errors together with time increasing backward can also be observed from Figures 11 and 12 where the standard deviations of the mean altitudes of mean trajectories increase while the time of the simulation increases.

When regarding both the cloud and aerosol cases it can be concluded that the FLEXPART mean trajectories are usually longer than Hysplit mean trajectories and that the differences between the two models are larger during cloud cases. In general these differences between the trajectories of the models can likely be explained by the differences in the altitudes of FLEXPART and Hysplit mean trajectories discussed in the Chapter 4.2.1. The explanation to why different altitudes cause differences in the locations of mean trajectories is associated with the different wind speeds and directions in different altitudes: If two particles are located in the same longitude and latitude, but at different altitudes, they may move in completely different directions or at completely different speeds due to variations of the wind with height.

Interesting characteristic can be found from the geographical locations of mean trajectories of cloud and aerosol cases. In cloud cases most of the trajectories come from South or South-West whereas in the aerosol cases most of the trajectories come from North or North-West. This indicates that in addition to the synoptic situation the source areas of the particles could be in the background causing different interesting observations, i.e. new particle formation and interesting cloud events.

5 Results from wet deposition studies

The aim of this chapter is to quantify the impact of wet deposition on how much mass of the particles is transported to SMEAR II (Hyytiälä, Finland). This aim is associated with the wet deposition parameterization of FLEXPART. The aim is studied by defining how much mass of the particles originates from the FLEXPART simulations (i.e. particles appearing during the simulation, schematic presentation in Fig. 2) and what are the origin altitudes of the mass of the particles.

Based on Figure 2, and its explanation in Chapter 2.1.2, some of the particles released at the beginning of the simulation physically originate from the area to which particles can spread over the 96 hour simulation (as the red particles in Fig. 2) and some particles physically originate outside of the area which particles can spread over the simulation (as the green particles in Fig. 2). The mass of the particles originating from the area which particles can spread, i.e. mass of the particles scavenged by wet deposition in backward in time simulation, is under research in this section. The mass of the particles originating from an altitude is defined by studying the altitudes where mass is scavenged in backward in time simulations.

The calculated amount of mass originating from the simulation and the origin altitude of the mass are normalized by the release mass, like explained in Chapter 3.2.2, to make the numbers easier for comparison. In this way the numbers are between zero (none of the particles originate from the simulation, i. e. none of the particles are removed in the simulation by wet deposition, or from the altitude) and one (the whole mass of the particles originate from the simulation, i. e. all of the mass of the particles is removed in the simulation by wet deposition, or from the altitude) and the dimension of the variable is kg/kg. For the particles which originate from the simulation the mean trajectory describes beside the path the origin area of the particles, while for the particles originating outside the simulation the mean trajectory describes only the path of the particles.

As explained in section 2.1.2, the effect of in-cloud scavenging is generally larger than the effect of below-cloud scavenging (Hertel et al., 1995; Webster et al., 2014). The differences between default below-cloud scavenging coefficient and smaller below-cloud scavenging coefficient are studied in this chapter to clarify, if the smaller effect of the below-cloud scavenging compared to the in-cloud scavenging can be observed in the FLEXPART simulations. If the dominant effect is in-cloud scavenging, there should not be a big difference in the masses originating from the simulation or the origin altitudes, or between the simulations conducted with different tracer particles (i.e. different below-cloud scavenging parameterizations).

Because the simulations analyzed in this chapter are conducted only with FLEXPART, the simulations are referred during this chapter only by *cloud case* and *aerosol case simulations* instead of *FLEXPART cloud case* and *FLEXPART aerosol case simulations*. The simulations analyzed in this chapter were conducted with WDT1 and WDT2 particles. WDT1 is a tracer particle which is influenced by the default below-cloud and in-cloud scavenging parameterizations. WDT2 particles are effected much less by below-cloud scavenging than WDT1 particles but the in-cloud parameterization is the same.

5.1 Mass originating from the simulations

5.1.1 Default below cloud scavenging

The left hand side of Table 3 shows the amount of mass originating from the cloud case simulations conducted with WDT1 and the right hand shows the amount of mass originating from the simulation in aerosol case simulations conducted with WDT1. In addition, the means and standard deviations of the cloud and aerosol cases are listed in Table 3. Based on the left hand side of Table 3 it can be seen that in the case 22.2.2014 the mass originating from the simulation is larger than the release mass. This larger mass is a result of rounding errors in the calculation method, calculation method is described in Chapter 3.2.2. More about the errors in the results are discussed in Chapter 6.

Table 3: *The amount of mass originating from the simulation in the simulations conducted with the wet deposition tracer of default below-cloud scavenging parameterization (WDT1). In addition, the means and standard deviations are listed.*

case	[kg/kg]	case	[kg/kg]
2.2.2014	0,99	8.4.2014	0,72
8.2.2014	0,99	22.4.2014	0,68
16.2.2014	0,88	23.4.2014	1,00
22.2.2014	1,01	24.4.2014	0,96
3.3.2014	0,95	25.4.2014	0,78
8.3.2014	0,97	26.4.2014	0,44
18.3.2014	0,97	27.4.2014	0,52
mean $\overline{x}_{cc,wdt1}$	**0,97**	**mean** $\overline{x}_{ac,wdt1}$	**0,73**
stand. dev. $s_{cc,wdt1}$	**0,04**	**stand. dev.** $s_{ac,wdt1}$	**0,21**

To verify that the means of the amounts of mass originating from the simulations are statistically different between the cloud and aerosol cases a statistical test is required. The most classic and best-known method for studying whether the means of two groups differ is called the two-sample Student's t-test (Wilcox, 2009). The goal of the two-sample Student's t-test is to test if the means of two populations are equal to each other:

$$H_0 : \overline{x}_1 = \overline{x}_2. \tag{5.1}$$

The t-test is made by calculating the t statistic and the when calculated t value is larger or equal to the T value of significance level α it can be said that the null hypothesis (Eq. 5.1) is rejected at α significance level. When the significance level α is smaller it is less likely that the means will be found to be the same. The most commonly used significance level α to prove that the means are different is 0,05.

The t statistic is calculated by

$$t = \frac{\overline{x}_1 - \overline{x}_2}{\sqrt{\frac{(n_1-1)s_1^2+(n_2-1)s_2^2}{n_1+n_2-2}}\sqrt{\frac{1}{n_1}+\frac{1}{n_2}}} \tag{5.2}$$

where $\overline{x}_1$ and $\overline{x}_2$ are the means and s_1 and s_2 are the standard deviations of the populations, n_1 and n_2 are sizes of the populations and $(n_1 + n_2 - 2)$ is degrees from freedom. In Table 4 there are listed the significance levels and T values when degrees of freedom is $(7 + 7 - 2) = 12$. In the studied FLEXPART case classes the size of population is always $n_1 = n_2 = 7$.

Table 4: *α significance levels and T statistics for two-sample Student's t-test when degrees of freedom is 12.*

	90%	50%	20%	10%	5%	2%	1%	0,5%
α	0,90	0,50	0,20	0,10	0,05	0,02	0,01	0,005
T	0,128	0,695	1,356	1,782	2,179	2,681	3,055	3,428

The t statistic between cloud case and aerosol cases, simulations conducted with WDT1 particles, is based on Equation 5.2 $t = 2,99$. Based on Table 4 the calculated value 2,99 is larger than the significance limit of $\alpha = 0,02$ (limit is 2,681) but smaller than the significance limit of $\alpha = 0,01$ (limit is 3,055) which conclude that the means of the amount of mass originating from the simulation in cloud cases and aerosol cases conducted with WDT1 are different at the 0,02 significance level.

In-cloud and below-cloud coefficients (Eq. 2.2 and 2.5) depend

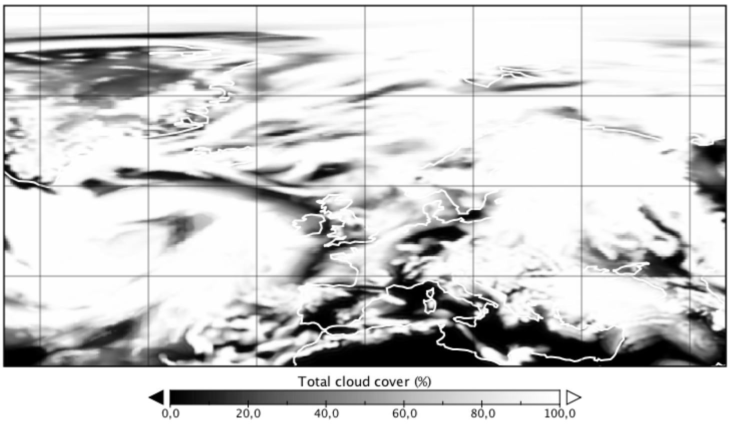

Figure 15: *Mean total cloud cover over Europe and Atlantic at 22.2.2014 0-6 UTC in NCEP GFS data.*

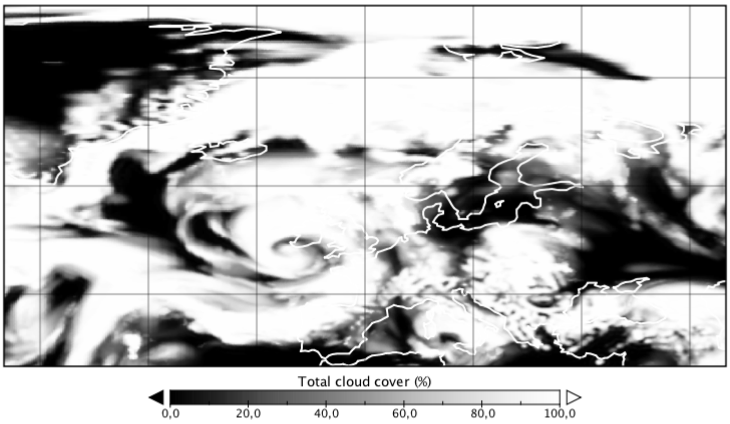

Figure 16: *Mean total cloud cover over Europe and Atlantic at 26.4.2014 0-6 UTC in NCEP GFS data.*

on subgrid scale precipitation, which again depends on precipitation rate and total cloud cover (Eq. 2.6). Whether, the in-cloud or below-cloud scavenging coefficient is used is based on the occurrence of cloud. Thus the three main factors that affect scavenging by wet deposition are precipitation, cloud cover and occurrence of cloud. Next the effect of cloud cover and occurrence of cloud on scavenging by wet deposition are discussed.

When calculating the scavenging by wet deposition the cloud fraction in a grid cell (F) is taken straight from the meteorological input data and the occurrence of cloud at some altitude is calculated based on the temperature and absolute humidity profiles in meteorological input data (Stohl et. al., 2010). The occurrence of cloud is assumed when the calculated relative humidity in a grid point is over 80%. If the humidity is over 80% and the occurrence of a cloud is assumed the in-cloud and below-cloud scavenging calculations are done in the grid cell, if the humidity is less than 80% the wet deposition in the grid cell is not calculated because the model assumes that there is not cloud.

To study the influence of humidity and cloudiness Figures 9 and 10 show the humidity profiles from the starting times of the cloud and aerosol case simulations and Figures 15 and 16 show the six hour mean total cloudiness in NCEP GFS data during the 0-6 UTC in the cases where the removed mass of the particles was largest (cloud case 22.2.2014) and smallest (aerosol case 26.4.2014) based on Table 3. NCEP GFS data is the data which was used as meteorological input data for FLEXPART simulations. The 80% humidity is marked on Figures 9 and 10 to represent the altitudes where clouds occur. Mean total cloudiness (Fig. 15 and 16) and humidity profiles (Fig. 9 and 10) do not represent the mean cloudiness and humidity profiles over the whole simulations but they give an indication of what the cloudiness was during the cases and whether the particles originate close to Hyytiälä.

In six of the seven soundings associated with the cloud case simulations, humidity is over 80% in the layer from the ground to the 2 kilometer altitude (Fig. 9). Also in the one case (the 5.3.2014 case) where humidity is less than 80% somewhere in the layer between 0–2 kilometers, the humidity is over 80% from ground to over 1,5 kilometers. Hence the below-cloud scavenging is not possible in cloud cases at least near the release time, because the calculated cloud in FLEXPART reaches the ground. Figure 15 shows that on the 22.2.2014 between 0-6 UTC the cloudiness was 100% almost everywhere in Finland. The mean trajectory of the 22.2.2014 case is short and it goes from Southern Finland to the Baltic Countries, Southern Sweden and Denmark (Fig. 13). In Figure 15 the cloudiness in the above mentioned areas is 100 %. The synoptic situation was quite similar over the whole simulation of the 22.2.2014 case and based on the similar synoptic situation it can be assumed that also the cloudiness was quite similar over the whole simulation of the 22.2.2014 case. High cloudiness and over 80% humidity in high layer might be the reasons why all of the mass of the particles originates from the simulation, i.e. is scavenged by wet deposition, in the 22.2.2014 case.

Only one humidity profile, associated with the aerosol case simulations, has humidity which exceeds 80 % (Fig.10, the 8.4.2014 case). Thus in the other six cases in-cloud or below-cloud scavenging can not occur at the times of the soundings. Clearly the humidity has to be over 80% somewhere in simulation so that there can be particles originating from simulation. The mean trajectory of aerosol case 26.4.2014 does a small loop over Southern Finland, Gulf of Bothia and Sweden and then goes over Southern Swedish and Norway and ends over the North Sea, near to the coast of Norway (Fig. 14). The cloudiness in the areas above varies between zero and 100 percent during 26.4.2016 0-6 UTC (Fig. 16), mostly being from zero to fifty percent. The amount of mass originating from the simulation was in the 26.4.2014 case 0,44. The low cloudiness and small humidity values match well to the amount of mass originating from the simulation.

Based on the cases 22.2.2014 and 26.4.2014, Figures 9 and 10 and Table 3 the amount of mass originating from simulation clearly depends on the occurrences of cloud, i.e. humidity, and total cloudiness. In the 22.2.2014 case the total cloud cover is high, the lower part of atmosphere is moist and all of the mass of the particles is originating from the simulation while in the 26.4.2014 case cloud cover is lower, the lower part of the atmosphere is dry and only half of the release mass originates from the simulation. Also based on Figures 9 and 10 the generally larger high humidity near the surface supports the theory that in-cloud scavenging is generally larger than below-cloud scavenging because of when the cloud base is in contact with the ground below-cloud scavenging can not occur.

5.1.2 Smaller below cloud scavenging

The left hand side of Table 5 shows the total amount of mass originating from the simulation in cloud case simulations conducted with WDT2 and the right hand side of Table 5 shows the total amount of mass originating from simulation in aerosol case simulations conducted with WDT2. To verify that the means of the aerosol and cloud cases are statistically different the t statistic is calculated, $t = 3,36$ (Eq. 5.2). Based on Table 4 the means of the mass of the particles originating from simulation differ between cloud cases and aerosol cases, conducted with WDT2, at 0,01 significance level.

Next statistical test were performed to determine if the means

Table 5: *Mass originating from the cloud and aerosol simulations conducted with wet deposition tracer particle of smaller below-cloud scavenging parameterization (WDT2). Also means and standard deviations are listed in the table.*

case	[kg/kg]
2.2.2014	0,88
8.2.2014	0,95
16.2.2014	0,78
22.2.2014	1,00
3.3.2014	0,92
8.3.2014	0,91
18.3.2014	0,83
mean $\overline{x}_{cc,wdt2}$	**0,89**
stand. dev. $s_{cc,wdt2}$	**0,08**

case	[kg/kg]
8.4.2014	0,58
22.4.2014	0,50
23.4.2014	0,91
24.4.2014	0,86
25.4.2014	0,38
26.4.2014	0,28
27.4.2014	0,13
mean $\overline{x}_{ac,wdt2}$	**0,52**
stand. dev. $s_{ac,wdt2}$	**0,29**

of the cloud and aerosol case simulations conducted with WDT1 are different compared to the means of the cloud and aerosol case simulations conducted with WDT2. The t statistic between cloud case simulations conducted with WDT1 and WDT2 is based on Eq. 5.2 $t = 2,14$ and the t statistic between aerosol case simulations conducted with WDT1 and WDT2 is based on Eq. 5.2 $t = 1,59$. Based on the calculated t statistics and the significance levels in Table 4 it can be said that the means of the mass of the particles originating from simulation in the cloud case simulations conducted with WDT1 and WDT2 are different at the 0,1 significance level and in the aerosol case simulation conducted with WDT1 and WDT2 the means are different at the 0,2 significance level. These significance levels are quite high and both are over the 0,05 significance level which is the most commonly used value to prove that the means are different. These high significance levels indicate that the simulations conducted with WDT1 and WDT2 do not differ considerably from each other. These results confirm our hypothesis: in-cloud scavenging parameterization plays a much larger role in wet deposition than below-cloud scavenging parameterization.

In the cloud case simulations conducted with WDT1 and WDT2 the difference between the means is $0,97 - 0,89 = 0,08$ and in aerosol case simulations the difference is $0,73 - 0,52 = 0,21$ based on Tables 3 and 5. The cloud case simulations are more statistically different, as proved above, than the aerosol cases because the standard deviation is very high between the aerosol case simulations conducted with

WDT2 (Tab. 5). The difference between the means in the cloud case simulations is likely small for two main reasons. Firstly, the small below-cloud scavenging does not affect the cloudiness in grid cells, which was, for example, very high in the 22.2.2014 case (Fig. 15) and potentially one reason for that all of the mass of the particles in the 22.2.2014 case come from the simulation. Secondly, there can't be below-cloud scavenging in situations where the humidity is over 80% near the surface, as was found in all of the soundings associated with the cloud case simulations (Fig. 9).

One case where below-cloud scavenging was larger than in-cloud scavenging can be found from the results. In aerosol case 27.4.2014 total amount of mass originating from the simulation conducted with WDT1 is 0,52 and in simulation conducted with WDT2 total amount of mass originating from the simulation is 0,13. These values mean that the amount of in-cloud scavenging was 0,13 and the amount of below-cloud scavenging was 0,39, if assumed that the amount off in-cloud scavenging was the same in both simulations, which however is not true (Chapter 5.2.2). Based on Hertel et al. (1995) situations where the effect of in-cloud scavenging is larger than the effect of below-cloud scavenging are really rare and usually associated with heavy rainfalls. During the time of the simulation of the 27.4.2014 case there were quite heavy rainfalls in northern parts of Finland, Sweden and Norway, which might be the reason for large below-cloud scavenging during the case.

5.2 Origin altitudes of the mass of the particles

5.2.1 Default below cloud scavenging

The results of this chapter are studied to attempt to identify at which levels in the atmosphere is wet deposition most efficient. Figures 17 and 18 show the altitudes where the mass of the particles originate in the simulations. The vertical resolution of the altitude grid is 50 meters as explained and justified in Chapter 3.3.2. Mass originating from the simulation in a specific case (Table 3) could be calculated by integrating over all of the altitudes.

In cloud case simulations all mass of particles originate from below 3,5 kilometers (Fig. 17). The largest layer where wet deposition

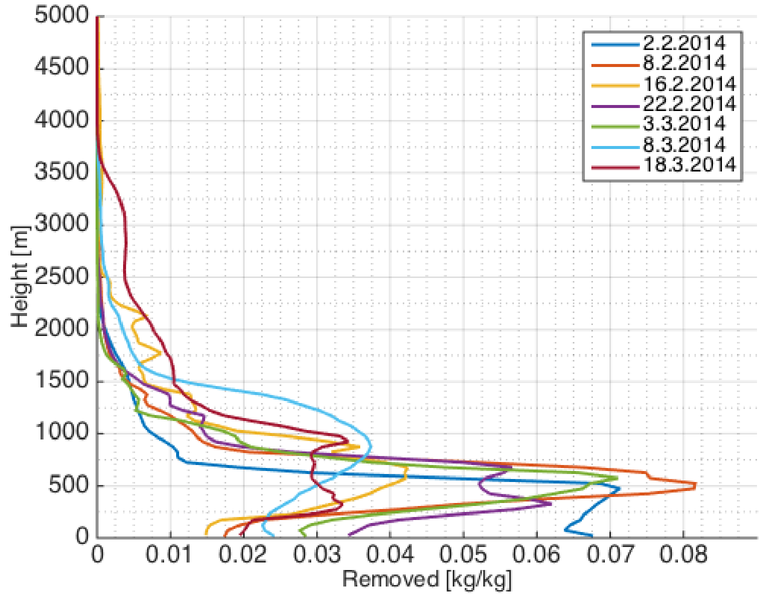

Figure 17: *Mass origin from a altitude in cloud case simulations conducted with WDT1 particles.*

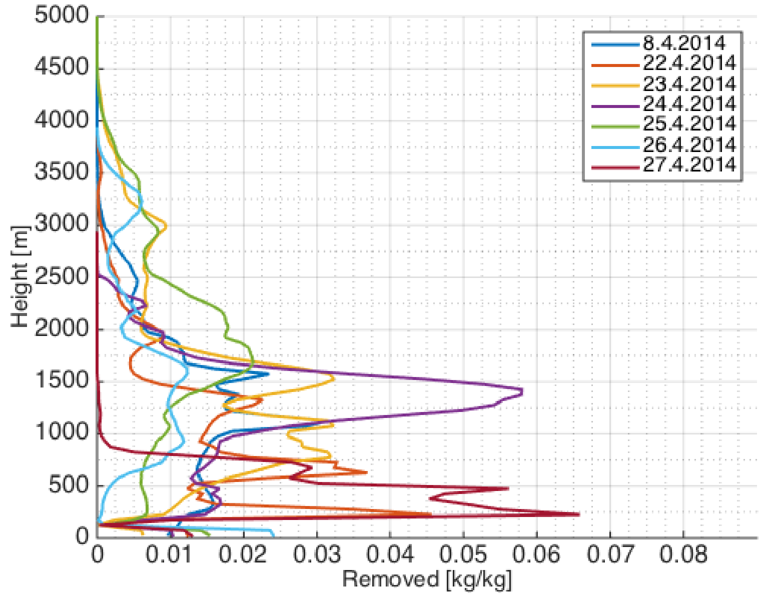

Figure 18: *Mass origin from a altitude in aerosol case simulations conducted with WDT1 particles.*

is active in cloud cases is between 250 and 1 000 meters. The largest peak in the amount of mass originating from an altitude in every cloud case simulations is within this range. The largest peak is in the 8.2.2014 case, where most of the mass of the particles originates from the layer between 500 and 550 meters. Less than 0,04 of the particles originate in six of the seven cases from near the surface. This factor indicates the small role below-cloud scavenging parameterization plays in the default parameterization.

In aerosol cases there is not a specific layer, as was found in the cloud case simulations, where most of the particles originate (Fig. 18). All of the particles originate below 4,5 kilometers, but most of the particles originate under 2 000 meters. Figure 10 shows that during the aerosol cases the humidity is generally highest near 1 500 meter altitude. High humidity near 1500 meter might be the explanation why the peak of mass originating from a altitude in the 23.4.2014 case is near 1 500 meters. This peak of 23.4.2014 is at the highest altitude of all of the cases.

The mass of the particles in cloud and aerosol case simulations generally originate from atmospheric boundary layer (ABL) in all of the cases, which indicates that wet deposition is most effective in ABL. Based on Stull (2015) ABL is a layer near to the Earth's surface where turbulence is ubiquitous and daily diurnal cycles of temperature, humidity and wind occur. The boundary layer height varies depending on the surface, cloudiness etc. but generally over the world the height of ABL is about 2 kilometers (Stuhl, 2015). The general characteristics of ABL varies between different regions and the 2 kilometer height is probably too high for Finland. Figure 10 suggest that the height of ABL is during most of the aerosol cases near 1,5 kilometers where the local maximum of humidity can be detected. There are lot of reasons why the particles generally originate from ABL. The most important reason is that the particles are released in boundary layer, in aerosol cases the release height was 100 meters and in cloud cases 0-500 meters. Turbulence in ABL mixes the particles to all over the boundary layer, does not transport particles easily to free troposphere. The reason for this is the inversion layer above the ABL (Stull, 2015), this inversion layer prevents the mixing with upper atmosphere.

Generally in all of the cloud and aerosol cases there is a clear local peak where particles are from a specific altitude instead of the origin of the particles being uniformly distributed with height. In aerosol cases the peaks aren't as large as in cloud cases but that can be a result of the smaller amount of mass originating from the simulations due to wet deposition in aerosol cases (Tab. 3). In aerosol cases the particles originate generally from higher altitudes than in cloud cases. This result is consistent with the generally higher altitudes of mean trajectories in aerosol cases than in cloud cases (Fig. 11 and 12).

5.2.2 Smaller below cloud scavenging

The results of this chapter are studied to identify at which levels in the atmosphere there are differences between the effects of default below-cloud and small below-cloud wet deposition parameterizations. Figure 19 shows the differences in the altitudes where the mass of the particles is originating in the cloud case simulations conducted with WDT1 and WDT2. Figure 20 shows the differences in aerosol case simulations. When the difference is negative more of the mass of the particles is from the altitude in the simulation conducted with WDT2 and when the difference is positive more mass is from the altitude in the simulation conducted with WDT1. If the amount of removed mass by in-cloud scavenging would be the same in both simulations the difference should be in zero everywhere else than where below-cloud scavenging took place. For example the case 8.2.2014 Figure 19 shows clearly that this is not the situation. This indicates that the particles that were removed by below-cloud scavenging in the simulations conducted with WDT1 can be removed by in-cloud scavenging in the simulations conducted with WDT2. This feature can flatten the differences between calculated masses originating from the simulation.

Generally the differences between simulations conducted with WDT1 and WDT2 are larger in aerosol cases than in cloud cases (Figures 19 and 20). This was the situation also in the total mass originating from the simulations and hence the results are consistent with each other (Tables 3 and 5). The only case (27.4.2014) where below-cloud scavenging was larger than in-cloud scavenging can be detected clearly from Figure 20. In the 27.4.2014 case the difference in mass originating from a specific altitude is larger than in any other case, being about 0,06 near the altitude of 250 meters. Altitude 225 meters is so low that it could be below-cloud altitude.

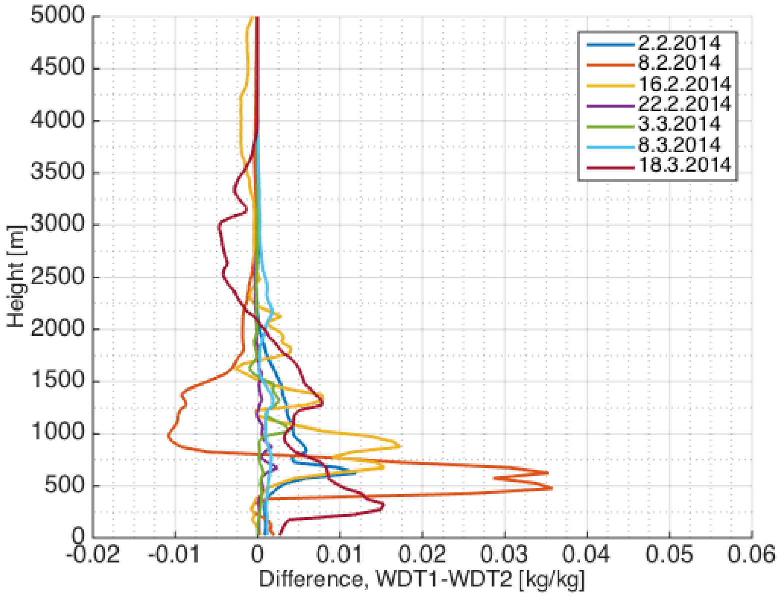

Figure 19: *Differences in mass originating from a altitude between cloud case simulations conducted with WDT1 and WDT2 particles.*

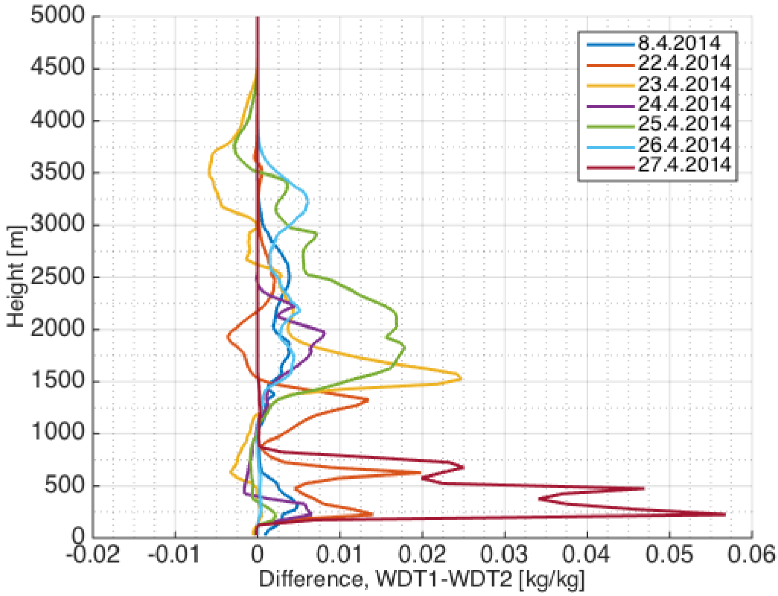

Figure 20: *Differences in mass originating from a altitude between aerosol case simulations conducted with WDT1 and WDT2 particles.*

6 Discussion

The results obtained in this study suggest that the particles detected at SMEAR II (Hyytiälä, Finland) during the cloud cases originate from a wider area than the particles detected during the aerosol cases. Also the results suggest that the effect of wet deposition is much larger in cloud cases than in aerosol cases. Furthermore, the results from the different wet deposition, in-cloud and below-cloud, parameterizations suggest that the in-cloud scavenging is the major removal process in both cloud and aerosol cases. In both cloud and aerosol cases classes the differences between default below-cloud scavenging and smaller below-cloud scavenging are not statistically different although in one of the aerosol cases the mass scavenged below-cloud was larger than the mass scavenged in-cloud.

This study differs considerably from almost all previous studies which have utilized FLEXPART and for that reason the results of the trajectory part of this study (Chapter 4) are important. When regarding the outputs of FLEXPART model, the concentration (forwards in time mode) and source-receptor relationship i. e. the potential source area (backward in time mode) are the mostly used and studied outputs and the trajectories of FLEXPART are less studied. The mean trajectories of FLEXPART are less studied, potentially because there are trajectory models like Hysplit, that require less calculation time and calculation space than FLEXPART. The comparison of the mean trajectories of FLEXPART is important because the comparison can show weaknesses in the model that FLEXPART is compared against or it can show problems in mean trajectories calculations of FLEXPART. Also when the different meteorological input data are used it can show how dependent the specific model is from the used meteorological data.

The differences in the FLEXPART and Hysplit simulations conducted for this thesis are mainly the result of two factors. The first factor is the differences in the assumptions and in the calculation methods of the models. Second factor is the different meteorological input data used for the simulations, in FLEXPART simulations the meteorological input data was NCEP CFS reanalysis data while in Hysplit the used meteorological input data was GDAS gridded observation data.

This study shows that the mean trajectories of FLEXPART and Hysplit are not identical, but this study does not take a position on which model is the best. There are differences in all of the studied features of trajectories: between the spread of FLEXPART plume trajectories and the spread of Hysplit ensemble trajectories, between the altitudes of mean trajectories and also in the geographical locations of the mean trajectories. The differences between the simulations of the models are more significant in cloud cases than in aerosol cases. As presented in the paragraph above the differences between the simulations of FLEXPART and Hysplit can be caused by the differences in the calculation methods of the models or by the different meteorological input data. Hegarty et al. (2013) compared FLEXPART and Hysplit with forward in time simulations that were compared with two controlled tracer release experiments. They suggested that the models are significantly sensitive to the meteorological input data and the differences in their calculation methods play a minor role and therefore supports the result presented in this thesis. The simulations that were used to study the differences between the models were made as equal as possible: wet deposition and other removal processes of FLEXPART were turned off and the release altitudes were different in cloud cases where the release height was 0-500 meters in FLEXPART and 100 meters in Hysplit. In aerosol cases the release height was 100 meters in both models. The results suggested that the release height is an important factor to the altitude of the mean trajectory. All these factors listed above may suggest that the more minor reason for the differences between the models was the differences in the calculation methods of the models. This result is in agreement with the result of Hegarty et al. (2013). The reason why the differences between the studied trajectory variables was smaller in aerosol cases than in cloud cases could be that the different meteorological input data of the models represent the high pressure situation more similarly than on low pressure situations. Also generally the pressure gradients and thereby the geostrophic winds are weaker in high pressure situations which may explain why in the high pressure systems the differences between meteorological fields may not be very large.

Currently there are no previous studies where the wet deposition parameterization of FLEXPART would be compared or evaluated against some tracer experiment or other model. Only one study that compared FLEXPART simulations with and without wet deposition and used FLEXPART in backward in time mode was found (Gadhavi et al., 2015). However, Gadhavi et al. (2015) did not even study clearly the effect of wet deposition, because they studied errors in the black carbon emissions which could be caused by other parameterizations in FLEXPART. This show clearly that to date there has been minimal evaluation of the wet deposition parameterization of FLEXPART.

As mentioned above the trajectories of FLEXPART are not fully studied before. For this reason the error associated with the paths of the trajectories can not be defined. Possibly the error is roughly the same, maybe little less than the error in traditional backward trajectories which is about 20 % of the length of the path that the air parcel has moved. In the calculated amounts of mass originating from the simulations and from the altitudes the errors can be quite large because of the wet deposition parameterization is not evaluated and also because of the rounding errors in the calculation methods.

Altogether the results from the trajectory studies and wet deposition studies support each other and in each variable that was studied the differences were larger within the cloud cases than within the aerosol cases. Generally aerosol cases can be described as cases where the mass of the particles originating from far away from the SMEAR II (Hyytiälä, Finland) and mass of the particles is originating from higher altitudes and the particles move with higher winds. In cloud cases most of the particles are originating from near SMEAR II (Hyytiälä, Finland) and the mass of the particles originates from lower altitudes, but not near the surface. In aerosol cases the impact of wet deposition on how much mass of the particles is transported to SMEAR II is smaller than in cloud cases.

7 Summary

There were two aims of this thesis, the first aim was to study the source areas of particles arriving to measurement station SMEAR II (Hyytiälä, Finland) represented by the Lagrangian particle dispersion model FLEXPART. The first aim included also comparison of FLEXPART results to results from traditional backward trajectory model Hysplit. The second aim was to study the impact of wet deposition on how much particles were transported to SMEAR II. Both of the aims were studied by using two classes of case studies and 14 case studies. The classes of case studies were aerosol and cloud cases and in both of them there were seven case studies. Aerosol case studies were selected based on interesting new particle formation cases and cloud cases were interesting cloud situations. For all of the case studies 96 hours backward in time simulations were made. To study the first aim, the FLEXPART simulations were made with passive tracer particles, and for the second aim the simulations were made with two different wet deposition tracer particles. To study the first aim also 96 hour simulations were made with trajectory model Hysplit.

To define the source regions of particles arriving to SMEAR II and to see how well FLEXPART can represent them, FLEXPART backward in time simulations were compared against backward in time simulations made with Hysplit. The comparison was made by comparing three variables: the spread of FLEXPART plume trajectories and Hysplit ensemble trajectories, the altitudes of FLEXPART and Hysplit mean trajectories and also by comparing the geographical locations of FLEXPART and Hysplit mean trajectories. In all of these three variables the differences between the models were larger in cloud cases than in aerosol cases, for example when comparing the mean altitudes of the mean trajectories the means of FLEXPART and Hysplit were not within the error bars in cloud case simulations while in aerosol case simulations they were mostly within the error bars. It was suggested that the reason for larger differences in cloud case simulations was that the release heights in FLEXPART and Hysplit cloud case simulations were different and aerosol case simulations the release heights were the same.

The second aim, the impact of wet deposition on how many particles were transported to Hyytiälä, was studied by defining how much of the mass of the particles is originating from FLEXPART backward in time simulations, i. e. how much of the mass of the particles is scavenged by wet deposition, and by identifying in what altitudes the mass originates from. When using the most commonly used significance level $\alpha = 0,05$, it can be concluded that the result was that statistically more mass originates from the simulations of cloud cases than of aerosol cases. For example, in the simulations where the default wet deposition parameterization was used in cloud cases 97 % of the particles were scavenged by wet deposition but only 73 % were in aerosol cases. To study whether the in-cloud scavenging is larger than the below-cloud scavenging, which was the hypothesis, two different sensitivity studies that had larger and smaller below-cloud wet deposition parameterization were done and the hypothesis was confirmed. The differences between the two sensitivity studies were not statistically different, in other words the contribution of below-cloud scavenging was really small and overall it did not affect the amount of wet deposition.

The conclusion for the first aim is that FLEXPART can represent the source areas of particles and meteorological input data has a significant effect on the path of the trajectories.The results also suggest that the release altitude significantly affect the spread of particles and their source areas. For the second aim, the result was that the impact of wet deposition on how many particles are transported to SMEAR II, Hyytiälä, is really important, especially in low pressure situations.

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Lasse deserves the biggest thanks because he is the one who supports me when I am not in my prime, $(x^2 + y^2 - 1)^3 - x^2 \times y^3 = 0$.

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A Correction factors

Correction factors used to calculate the area fraction which experiences precipitation (equation 2.7). Depending on the factor precipitation rate I refers to large scale precipitation rate I_l or convective precipitation rate I_c. Units of precipitation rates are in mm/hour.

Factor	$I<1$	$1<I\leq 3$	$3<I\leq 8$	$8<I\leq 20$	$20<I$
fr_l	0,50	0,65	0,80	0,90	0,95
fr_c	0,40	0,55	0,70	0,80	0,90

B Technical details of FLEXPART simulations

The example option files presented below are from the 26.4.2014 case. Only COMMAND, OUTGRID and RELEASE files are presented because they were the only files which options were changed.

The option file called COMMAND:

```
*******************************************************************************
*                                                                             *
*      Input file for the Lagrangian particle dispersion model FLEXPART       *
*                        Please select your options                           *
*                                                                             *
*******************************************************************************

1. --                  3X, I2
   -1
   LDIRECT             1 FOR FORWARD SIMULATION, -1 FOR BACKWARD SIMULATION
2. -------- ------     3X, I8, 1X, I6
   20140422 080000
   YYYYMMDD HHMISS     BEGINNING DATE OF SIMULATION
3. -------- ------     3X, I8, 1X, I6
   20140426 080000
   YYYYMMDD HHMISS     ENDING DATE OF SIMULATION
4. -----               3X, I5
    3600
   SSSSS               OUTPUT EVERY SSSSS SECONDS
5. -----               3X, I5
    3600
   SSSSS               TIME AVERAGE OF OUTPUT (IN SSSSS SECONDS)
6. -----               3X, I5
     600
   SSSSS               SAMPLING RATE OF OUTPUT (IN SSSSS SECONDS)
7. ---------           3X, I9
   999999999
   SSSSSSSSS           TIME CONSTANT FOR PARTICLE SPLITTING (IN SECONDS)
8. -----               3X, I5
```

```
      600
    SSSSS              SYNCHRONISATION INTERVAL OF FLEXPART (IN SECONDS)
9.  ---.--             4X, F6.4
     -2.0
    CTL                FACTOR, BY WHICH TIME STEP MUST BE SMALLER THAN TL
10. ---                4X, I3
      4
    IFINE              DECREASE OF TIME STEP FOR VERTICAL MOTION BY FACTOR IFINE
11. -                  4X, I1
    5
    IOUT               1 CONCENTRATION OUTPUT, 2 MIXING RATIO OUTPUT, 3 BOTH,4 PLUME TRAJECT., 5=1+4
12. -                  4X, I1
    0
    IPOUT              PARTICLE DUMP: 0 NO, 1 EVERY OUTPUT INTERVAL, 2 ONLY AT END
13. _                  4X, I1
    0
    LSUBGRID           SUBGRID TERRAIN EFFECT PARAMETERIZATION: 1 YES, 0 NO
14. _                  4X, I1
    0
    LCONVECTION        CONVECTION: 1 YES, 0 NO
15. _                  4X, I1
    0
    LAGESPECTRA        AGE SPECTRA: 1 YES, 0 NO
16. _                  4X, I1
    0
    IPIN               CONTINUE SIMULATION WITH DUMPED PARTICLE DATA: 1 YES, 0 NO
17. _
    1                  4X,I1
    IOFR               IOUTPUTFOREACHREL CREATE AN OUPUT FILE FOR EACH RELEASE LOCATION: 1 YES, 0 NO
18. _                  4X, I1
    0
    IFLUX              CALCULATE FLUXES: 1 YES, 0 NO
19. _                  4X, I1
    0
    MDOMAINFILL        DOMAIN-FILLING TRAJECTORY OPTION: 1 YES, 0 NO, 2 STRAT. O3 TRACER
20. _                  4X, I1
    1
    IND_SOURCE         1=MASS UNIT , 2=MASS MIXING RATIO UNIT
21. _                  4X, I1
    1
    IND_RECEPTOR       1=MASS UNIT , 2=MASS MIXING RATIO UNIT
22. _                  4X, I1
    0
    MQUASILAG          QUASILAGRANGIAN MODE TO TRACK INDIVIDUAL PARTICLES: 1 YES, 0 NO
23. _                  4X, I1
    1
    NESTED_OUTPUT      SHALL NESTED OUTPUT BE USED? 1 YES, 0 NO
24. _                  4X, I1
    0
    LINIT_COND         INITIAL COND. FOR BW RUNS: 0=NO,1=MASS UNIT,2=MASS MIXING RATIO UNIT
```

The option file called OUTGRID:

```
********************************************************************************
*                                                                              *
*      Input file for the Lagrangian particle dispersion model FLEXPART        *
*                       Please specify your output grid                        *
*                                                                              *
********************************************************************************

1.  ------.----         4X,F11.4
       -23.0000         GEOGRAFICAL LONGITUDE OF LOWER LEFT CORNER OF OUTPUT GRID
    OUTLONLEFT          (left boundary of the first grid cell - not its centre)
2.  ------.----         4X,F11.4
```

```
     50.0000          GEOGRAFICAL LATITUDE OF LOWER LEFT CORNER OF OUTPUT GRID
   OUTLATLOWER        (lower boundary of the first grid cell - not its centre)
3. -----              4X,I5
     751              NUMBER OF GRID POINTS IN X DIRECTION (= No. of cells + 1)
   NUMXGRID
4. -----              4X,I5
     201              NUMBER OF GRID POINTS IN Y DIRECTION (= No. of cells + 1)
   NUMYGRID
5. ------.---         4X,F10.3
        0.100         GRID DISTANCE IN X DIRECTION
   DXOUTLON
6. ------.---         4X,F10.3
        0.100         GRID DISTANCE IN Y DIRECTION
   DYOUTLAT
10. -----.-           4X, F7.1
     100.0
   LEVEL 4            HEIGHT OF LEVEL (UPPER BOUNDARY)
10. -----.-           4X, F7.1
    5000.0
   LEVEL 4            HEIGHT OF LEVEL (UPPER BOUNDARY)
10. -----.-           4X, F7.1
   10000.0
   LEVEL 4            HEIGHT OF LEVEL (UPPER BOUNDARY)
```

The option file called RELEASES:

```
************************************************************************
*                                                                      *
*                                                                      *
*                                                                      *
*   Input file for the Lagrangian particle dispersion model FLEXPART   *
*                        Please select your options                    *
*                                                                      *
*                                                                      *
*                                                                      *
************************************************************************
+++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++
  1
---                        i3    Total number of species emitted
  1
---                        i3    Index of species in file SPECIES
=========================================================================
20140426 080000
-------- ------            i8,1x,i6 Beginning date and time of release
20140426 080000
-------- ------            i8,1x,i6 Ending date and time of release
  24.270
----.----                  f9.4  Longitude [DEG] of lower left corner
  61.840
----.----                  f9.4  Latitude [DEG] of lower left corner
  24.290
----.----                  f9.4  Longitude [DEG] of upper right corner
  61.860
----.----                  f9.4  Latitude [DEG] of upper right corner
        1
---------                  i9    1 for m above ground, 2 for m above sea level
  100.00
-----.---                  f10.3 Lower z-level (in m agl or m asl)
  100.00
-----.---                  f10.3 Upper z-level (in m agl or m asl)
    50000
---------                  i9    Total number of particles to be released
1.5000E01
-.----E--                  e9.4  Total mass emitted
```

RELEASE_26042014

```
----------------------------------------    character*40  comment
++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++++
```