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New method to quantify contribution to aerosol particles from new particle formation: Comparison with traditional method at Vavihill background field station in Sweden

Danielle Appavoo and Kristine Confalone

June 21, 2015

Abstract

Aerosol particles exist in the atmosphere in large concentrations and have significant global climate effects. New particle formation is an aerosol particle source and, to date, its contribution to the total aerosol concentration in the atmosphere is not fully understood. This study focuses on the extension of new particle formation events that begin in Lille Valby, Denmark and travel over 70 km to Vavihill, Sweden. Data over a five year period (2005, 2006, 2008, 2009 and 2010) was analyzed. Dates selected for analysis had to have a new particle formation event in Lille Valby and be either undefined, have an unclear event, or have a non-event in Vavihill as defined by traditional methods from Dal Maso et al. (2005). The growth rate, particle size distributions, and wind trajectories at the time of the event were considered when analyzing each event case. 7 out of 1480 days met all of the criteria to be classified as new particle formation extended from Lille Valby. If the result is extrapolated to all wind directions and sources, it is likely that a much higher percentage of the days would meet the criteria. The average percentage of particles in Vavihill that can be attributed to new particle formation from Lille Valby on reclassified days at event times over the five year period considered is 64.18%.

1. Introduction

Aerosol particles are made up of solid and liquid particles suspended in a gas. Although not fully understood, aerosol particles are known to have many effects on climate including the scattering and absorption of solar radiation as well as increasing the number of cloud droplets as they act as cloud condensation nuclei. New particle formation is a key concept to this study. A new particle formation event is classically defined as a distinct mode in the particle size distribution beginning in the nucleation mode size range that shows signs of growth over time. These events often take the shape of a “banana” when particle number size distributions are plotted. Figure 1 below shows a new particle formation event in Vavihill illustrating the “banana” shape. When this data is examined, if there are particles within the $<25\text{nm}$ size range but no growth is observed, it is classified as a non-event. As defined by Dal Maso et al. (2005) undefined events are those which cannot be classified as an event or a non-event. When data collected over five years was inspected, approximately 38.34% of days in Vavihill, Sweden are classified as undefined or unclear events and 51.23% are classified as non-events. Understanding the origin of these non-event, undefined, and unclear events could help give insight to the climate effects of newly formed aerosol particles.

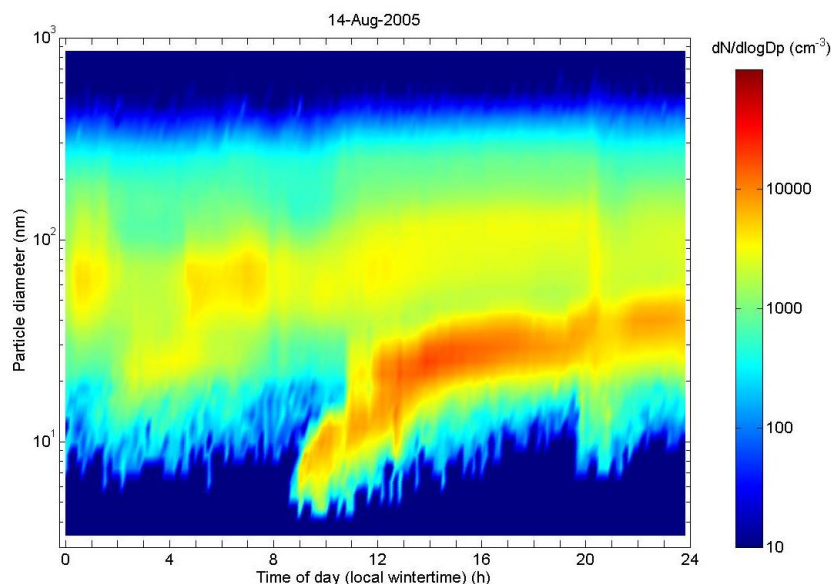


Figure 1. Size distribution data showing a new particle formation event in Vavihill on August 14, 2005

2. Objectives

The specific aims of the project are:

1. To re-classify previously undefined events in Vavihill as new particle formation from a nearby station in Lille Valby, Denmark.
2. To determine the number of previously unclassified aerosol particles detected in Vavihill that can be related to new particle formation events in Lille Valby, Denmark.

3. Background

3.1 Definition of an Aerosol

An aerosol is a mixture of solid and liquid particles which are suspended in gas. They are always present in the atmosphere despite having extremely variable concentrations and a short residence time. Aerosol particles are microscopic in size and therefore can only be seen when their concentrations are large enough. This often occurs in the form of haze, fog or smoke. They can also be detected when they are deposited onto the earth in a large quantity, such as with dust particles in the desert.

Aerosols vary greatly by size and composition in the atmosphere. Therefore it is beneficial to analyze a population of aerosols as opposed to individual particles. Some important characteristics of a population of aerosols includes size distribution, chemical compositions and shape of the particles.

Aerosols have a wide range of both natural and anthropogenic sources. The driving force in the formation of aerosols varies but several important ones are wind, friction, combustion, and nucleation. Primary aerosols are those emitted directly into the atmosphere, such as those produced through combustion or due to wind friction on various surfaces. Secondary aerosols are particles which originate from the condensation of atmospheric gas-phase species.

3.2 Sources of Aerosols

Aerosols come from a variety of both natural and anthropogenic sources. Primary sources, secondary sources, and nucleation or new particle formation (NPF) will be discussed.

Primary particle sources include mechanical generation as well as combustion generation. Friction is the main mechanical process by which aerosols are generated. Wave breaking generates aerosols from sea spray (marine aerosols) when wind friction lifts small particles of salt water from the wave. Primary biogenic aerosols also travel via the wind. These aerosols are insect and plant particles (i.e. pollen, spores, bacteria, viruses) of varying sizes. Wind friction also generates desert dust aerosols. Soil particles are lifted off the Earth's surface as wind travels over large flat areas such as the desert. Friction between the wheels of a vehicle and a surface also generates aerosols. This source is called vehicle abrasion. (Boucher, 2012)

Combustion sources of primary particles have natural and anthropogenic origins. Volcanic eruptions are an example of a natural source. Volcanic aerosols are generated from volcanic ash particles that travel large distances. Sulphur dioxide released from volcanic eruptions can also proceed to form secondary aerosols. Combustion of biomass is both a natural and anthropogenic source. The burning of any biological material, excluding fossil fuels, falls under this classification. Forest fires and domestic wood combustion are natural sources of these aerosols. Biomass

combustion releases organic carbon based compounds as well as gaseous compounds that contribute to generating secondary aerosols. The combustion of fossil fuels is an anthropogenic aerosol source. The burning of natural gas and coal produces black and organic carbon. It also produces sulphur dioxide which acts as a precursor to secondary aerosols. (Boucher, 2012)

Gas to particle conversion can be divided into two main categories; secondary particle formation and new particle formation. As previously stated, secondary particle gaseous precursors can come from volcanic eruptions, biomass combustion, and fossil fuel combustion. There are also secondary aerosols that come from biogenic origins such as phytoplankton, a type of oceanic biota which produces dimethyl sulfide. This compound along with other volatile organic compounds emitted from plants and algae act as secondary particle precursors. (Boucher, 2012)

Small, new particles are formed by isoprenes and monoterpenes released by forests as they are exposed to sunlight. This phenomenon is called new particle formation. New particle formation (or nucleation) as an aerosol source is the focus of this study and is responsible for much of the aerosol concentration in the atmosphere. Nucleation is the transformation of gas molecules into a cluster of molecules forming an aerosol nuclei (approximately 1 to 2 nanometers in diameter). Particles below 3 nanometers in diameter are referred to as clusters. Five theories are considered as processes for nucleation; two heterogeneous and three homogeneous. Heterogeneous nucleation involves gas molecules interacting with preexisting clusters of small particles which have different chemical compositions. The types of heterogeneous nucleation are cluster activation and ion-induced nucleation. Homogeneous nucleation is the interaction between molecules of the same type to form particle clusters (aerosol nuclei). The types of homogeneous nucleation are kinetically controlled nucleation, binary water sulfuric acid nucleation and ternary water sulfuric acid ammonia nucleation. (Boucher, 2012)

3.3 Chemical Composition

Due to the varying sources of aerosols as described above, their chemical composition can change based on their environment. Chemical composition is a key factor in the aerosols ability to take up water and grow in size, therefore affecting their role as cloud condensation nuclei. An aerosol mixture can be either external or internal. In an external mixture each particle is chemically pure so the mixture contains particles of different compositions. In this case a different size distribution can be made for each aerosol type in the population. In an internal mixture each particle is a mix of different chemical species. If it is a perfect mixture then each particle has the same chemical composition and the population can be characterized by a single distribution. The particles themselves can be made up of many different species such as black carbon, ammonium sulphate, sodium chloride and organics. (Boucher, 2012)

In this study, the focus is on new particle formation within boreal forests. Some of the aerosols emitted in these environments come from vegetation and can include spores, fungi and leaf matter. These are known as primary biological aerosol particles. However a more prominent and well known effect of these forests is the release of biogenic volatile organic compounds (BVOCs) which

are also emitted by vegetation. Once released into the atmosphere these BVOCs can be oxidized to form products that condense onto existing aerosol particles, creating secondary organic aerosol (SOA). (Spracklen et al. 2008)

3.4 Atmospheric Processes

Once emitted into the atmosphere, aerosols undergo a number of atmospheric processes. These processes dictate the particles' residence time in the atmosphere due to their effect on the chemical composition and size of the aerosols.

Evaporation and condensation describe how much mass is transferred from the gaseous phase to the particulate phase and vice versa. Consider a single aerosol droplet in the atmosphere. Gas-phase compounds can condense onto this droplet just as condensed matter can evaporate back into the gaseous phase. This mass transfer is driven by a concentration gradient between the gas phase far from the aerosol and at the aerosol surface. At equilibrium, the condensation rate and the evaporation rate are the same due to equal concentrations inside the aerosol droplet and in the gas phase just outside the droplet. A difference in concentration in the gaseous phase disturbs this equilibrium and allows for either condensation or evaporation to dominate. (Boucher, 2012)

Condensation is very important for aerosols in the accumulation mode as it allows growth from diameters of a few nanometers up to one micrometer. It is a function of the vapour pressure of the gas phase species, its saturation vapour pressure, the curvature radius of the particle and the affinity between the gas and the particle.

Coagulation is the aggregation or collection of aerosol particles to form larger particles. It occurs when particles are set in motion due to collisions with other fast moving molecules or particles. This is also known as Brownian motion. This process is very important for growth when large concentrations are present, for example in a polluted environment with small particles. In the troposphere, coagulation works to increase the mixing of aerosols and in the stratosphere it facilitates the growth of accumulation mode particles. (Boucher, 2012)

Aerosols also act as condensation sites for water in the atmosphere. Water vapour can condense onto a hydrated aerosol particle to become a cloud droplet. Cloud droplets then continue to grow in size as they collide with each other in the atmosphere. Aerosols which are not already part of a cloud droplet can coagulate with existing cloud droplets in the same way. If the cloud formed does not produce precipitation due to meteorological conditions, the cloud droplets will evaporate back into the atmosphere. When this happens the cloud condensation nuclei or atmospheric particle they nucleated upon will be released along with all soluble and insoluble material they collected. The effect of aerosols acting as cloud condensation nuclei has to do with how much light is scattered in the atmosphere. An increase in concentration of atmospheric particles increases the number concentration of cloud droplets. This creates a larger surface area to scatter light from the sun, therefore increasing cloud albedo. (Boucher, 2012)

Wet deposition or wet scavenging is the main sink of aerosols in the atmosphere. It occurs in clouds during the formation of precipitation and under clouds due to the downward flux of precipitating water. When clouds form in the atmosphere some aerosols get trapped in the aqueous phase as cloud condensation nuclei or through impaction by colliding with a cloud droplet. When these cloud droplets grow to a considerable size with a large sedimentation velocity, precipitation occurs. Through this precipitation aerosols are removed from the atmosphere when they reach the Earth's surface. This is known as in-cloud scavenging. Below-cloud scavenging occurs when the precipitation collides with atmospheric particles as it is falling, bringing them too to the Earth's surface. However, this process can be affected if precipitation is re-evaporated into the atmosphere as it is falling. This redistributes the aerosol mass back into the atmosphere. (Boucher, 2012)

Dry deposition is another sink of aerosol mass that occurs in areas with little to no precipitation. It refers to the direct deposit of aerosols back onto the Earth's surface. This occurs due to a turbulent flux of aerosols when they are relatively small. Aerosols can also be deposited onto the surface of the Earth through fog or mist which lie close to the ground. As with wet deposition, particles which have been brought to the Earth through dry deposition can be re-suspended into the atmosphere creating pollution and/or dry regions. (Boucher, 2012)

Sedimentation has a similar effect on aerosols as dry deposition, however it deals with particles of a larger size. Also known as gravitational settling it creates an aerosol sink at the surface of the Earth as well as redistributes aerosols vertically in the atmosphere. While sedimentation is not dominant in the troposphere it is very important in the stratosphere where vertical transport is slow. Here it works to bring aerosols down into the troposphere where they can be transported by large-scale transport, convective transport or wet scavenging. However, in the troposphere sedimentation does help to bring coarse aerosols back down to the surface after they have been emitted. How this occurs is governed by the sedimentation velocity which is determined through the difference between the gravitational and drag forces acting on the particles. (Boucher, 2012)

3.5 Climate Effects

Aerosols have a great effect on Earth's climate through their interaction with the atmosphere, the cryosphere, the biosphere and the ocean. It is understood that anthropogenic aerosols have counterbalanced the full warming effect of greenhouse gases on the atmosphere for many years. However the extent of this effect is still unknown. What is known is that atmospheric aerosols affect Earth's climate and radiative balance in three main ways.

The first effect is through the scattering and absorption of solar radiation. Aerosols scatter radiation from the sun back towards space, reducing the amount that reaches Earth's surface. They also absorb radiation, which heats the aerosol layer but like scattering results in cooling at the Earth's surface. This is known as the aerosol direct effect. The aerosol semi-indirect effect also has to do with the absorption of solar radiation, but refers to its' influence on the vertical temperature profile. This effect impacts meteorological parameters such as relative humidity, atmospheric stability and cloud formation. (Boucher, 2012)

The two remaining effects are indirect. As previously mentioned atmospheric aerosols act as cloud condensation nuclei, facilitating the formation of cloud droplets. This means that an increase in aerosols can result in an increase in cloud droplets. With this increase comes a decrease in size of cloud droplets as well as an increase in cloud reflectivity. Higher reflectivity means that less solar energy is absorbed, resulting in cooling of the climate system. This is known as the first indirect effect. Smaller cloud droplets means that it is more difficult for the cloud to initiate precipitation, therefore affecting the evolution of the cloud. This is known as the second indirect effect. (Boucher, 2012)

Radiative forcing refers to the disruption of the Earth's radiation energy budget by some activity. This energy balance is negatively affected by aerosols due to their overall cooling effect on the climate. Their cooling effect and thus their impact on the radiation energy budget can be calculated by quantifying their effects as cloud condensation nuclei. (Kurten et al., 2003)

4. New Particle Formation

4.1 Introduction of New Particle Formation Characteristics

Aerosol formation and growth is observed at many different stations around the world and, according to Kulmala et al. (2004), occurs nearly everywhere from urban plumes to coastal environments to clean arctic air to the free troposphere. In several different test locations, it was reported that the growth rates were similar with the highest growth rates of new particles being in the summer and lowest in the winter. The frequency of new particle formation events varies both seasonally and with location. In the Hyytiälä station in Finland, the highest frequency of new particle formation events occurs between March and May with another lower peak in September. In the Hohenpeissenberg station in Germany the highest frequency of new particle formation events occur in spring and winter. In St. Louis, U.S. no clear peaks in frequency are observed however the lowest number of new particle formation events is observed in winter. New particle formation events occur globally and vary due to the time of year and location. Methods to classify new particle formation events to further understand their patterns are discussed below.

4.2 Traditional Classification as Described by Dal Maso

A comprehensive methodology for classifying new particle formation events developed by Dal Maso et al. (2005) was used to analyze data in this study. This method was intended to be applicable for many sites worldwide and was in fact tested by Kristensson et al. (2008) and found to apply to southern Scandinavian conditions. Therefore this classification methodology was determined appropriate for the two sites considered in this study.

The criteria used to classify a new particle formation event described by Dal Maso et al. (2005) is summarized as:

1. A distinctly new mode of particles must appear in the size distribution
2. The mode must start in the nucleation mode size range
3. The mode must prevail over a time span of hours
4. The new mode must show signs of growth

Data was analyzed visually on a day to day basis by a four person group to minimize the effect of subjective opinions. The above criteria were used to distinguish event days from nonevent days as well as between new particle formation and point sources (caused by local pollution). Any disagreement within the panel regarding event/nonevent days resulted in the day being classified as undefined. This often occurs when particles appear sporadically in the nucleation size mode range or a later phase of a mode appears growing in the Aitken mode size range. Additionally any day with bad or missing data that affected the classification of the event resulted in the day being classified as “bad/missing data”.

4.3 New Classification as Described by Buenrostro

Buenrostro et al. (2009) proposes a framework to classify days that were previously unclassified by the classification set out by Dal Maso et al. (2005). The goal was to determine what characteristics undefined and event days had in common that prevented them from being classified as non-event days and also to identify qualities of the undefined days that the non-event days lacked. Data from event, non-event, and undefined days from 2004 at Hyytiälä was studied to develop the new classifications.

Five new classifications were arrived at; failed events, pollution related concentration peaks, ultrafine mode peaks, unclassified, and missing data. Failed events are further classified as quasi events and tail events. Quasi events are defined as nucleation particles (diameter ranging from 3-10 nm) that fail to achieve clear growth and/or lasts for less than one hour. Tail events are instances of nucleation elsewhere that are brought by the wind to the sample location. They are defined as a mode that appears at a diameter greater than 10 nm and grows over a period of several hours. Pollution related concentration peaks are classified as peaks that coincide with periods of high SO₂ or NO_x concentrations. Ultrafine-mode peaks are further classified into Aitken-mode peaks and nucleation-mode peaks. Aitken-mode and nucleation-mode peaks are those with no growth that exist in the 10-100 nm and 3-20 nm size ranges respectively. They also must correspond with low SO₂ and NO_x concentrations. Unclassified dates are those that do not fit with any of the previously listed classifications.

4.4 Extension of NPF

Several other studies on the extension of new particle formation over a region have been conducted. Hussein et al. (2009) has shown that new particle formation mode particles from events in southern Finland can be defined as a source of accumulation mode particles over Scandinavia.

Kristensson et al. (2014) developed a method using trajectories and size distribution data from a specific field site to estimate the spatial distribution of NPF around the measurement station. This method, referred to as the NanoMap method is able to pinpoint regions with a high or low number of NPF events.

This extension of new particle formation is the main interest of this study. Little is known about the travel of aerosol particles and how this affects areas near regions with a higher number of NPF events. These previous works show that this information is important in order to understand the extent to which newly formed particles affect climate on a local and global scale.

5. Method

5.1 Description of Data Collection Sites

The two locations that are the focus of this study are Vavihill in Sweden and Lille Valby in Denmark, shown in Figure 1. Both regions are forested areas where new particle formation events have been observed. Data collected at these sites over five years (2005 through 2010 excluding 2007) is used for this study.



Figure 2. Map of data collection sites obtained from Google Maps.

5.2 Instrumentation

This section outlines the measurement techniques employed in this project. The instruments used includes a differential mobility particle sizer at the Vavihill station and a scanning mobility particle sizer at the Lille Valby station.

5.2.1 Differential Mobility Particle Sizer (DMPS)

Measurements taken at the Vavihill station were done so using a differential mobility particle sizer (DMPS) system. This system was designed, constructed and calibrated at Lund University. The sampled air enters the system through a PM10 inlet and goes first into the bipolar charger. Here the particles are charged according to a well defined charge distribution. The flow is then split and put through two different differential mobility analyzers (DMAs). Inside these DMAs the particles are selected according to their size or electric mobility by an electric field. The first flow goes through a Nano-DMA which scans particles from 3.4 to 21.5 nm in diameter. Simultaneously the second flow goes through a DMA which scans particles from 21.5 to 857 nm in diameter. The two DMAs step in

equal logarithmic diameter intervals giving 37 mobility channels and a measure time of 10 minutes for one full number size distribution.

The two flows continue into two condensation particle counters (CPCs) which detect and count any mobility classified particles exiting the DMAs. The flow from the Nano-DMA goes into model CPC 3025 and the flow from the DMA goes into model CPC 7610 (TSI Inc., Shoreview, MN, USA). The measured mobility distributions are then converted into particle size distributions through the use of an algorithm for inversion of the DMPS data. (Kristensson et al., 2008)

5.2.2 Scanning Mobility Particle Sizer (SMPS)

Measurements taken at the Lille Valby station were done so using a scanning mobility particle sizer (SMPS). An SMPS consists of a drier, a bipolar charger, a differential mobility analyzer and a condensation particle counter. The air enters the SMPS and goes through the drier before entering the bipolar charger. As mentioned above, inside the bipolar charger aerosols are charged to a known charge distribution and then enter the DMA with this new charge distribution. Just as in the DMPS system, particles are classified according to their electrical mobility inside the DMA and then enter the CPC which detects and counts the classified particles. (Malmborg, 2014)

5.3 New Method

5.3.1 Selection of Cases

Cases for analysis were selected from five years of previously classified data (using the Dal Maso method). Each day, the data from Lille Valby or Vavihill was classified as either -2, -1, 0, 1, or 2. The definition of each of these classes is shown below in Table 1. Cases selected for the possibility of reclassification had to be a type 1 in Lille Valby and a type -1, 0 or 2 in Vavihill.

Table 1. *Traditional Classifications*

Classification	Definition
-2	Bad data or missing data
-1	Non-event
0	Undefined
1	Clear event
2	Unclear event

5.3.2 Wind Trajectory

The wind trajectory for each day selected was analyzed to determine whether the case was plausible or not. If it was found that the wind reaching Vavihill had passed over or near Lille Valby it was kept for further analysis. If the wind trajectory revealed that the air had come from another location, such as from the north of Sweden or across the Baltic Sea without crossing Denmark, the case was removed from the selection.

In order to perform this analysis, meteorological data was taken from the National Oceanic and Atmospheric Administration (NOAA) including wind speed and direction across the vertical plane in the atmosphere. Wind trajectories were then generated using the HYSPLIT trajectory model from NOAA. An image was created that showed where the wind was located on an hourly basis prior to reaching Vavihill. See Figure 3 below for an example of a generated image. Therefore at any given time in the day, it could be observed where the wind was located any number of hours before. This data was used to determine whether or not the particles found in Vavihill had travelled from Lille Valby within the correct time frame.

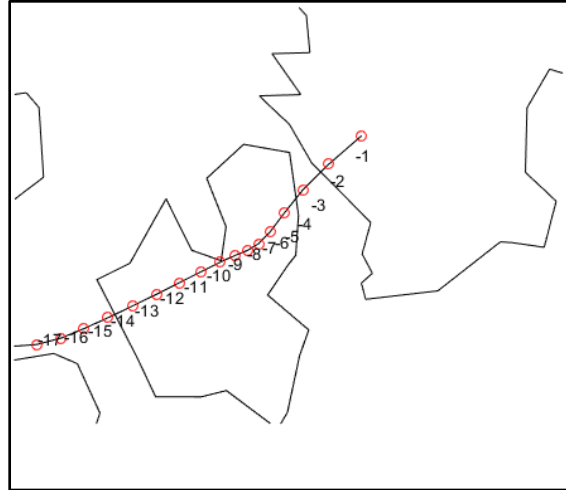


Figure 3. Image showing an ideal wind trajectory taken from May 12, 2006 at 12:00am.

5.3.4 Growth Rate Calculation

By analyzing the size distribution data from the Lille Valby station, the growth rate of each new particle formation event was calculated. This was done by performing a linear approximation using the time the particle formation began and ended as well as the initial and final sizes of the particles. See Figure 4 below for an example of the linear approximation. By comparing these two parameters a growth rate of particle size per hour was determined.

After the growth rate for each event was calculated, the subsequent event in Vavihill had to be analyzed to ascertain whether the two particle formation events matched.

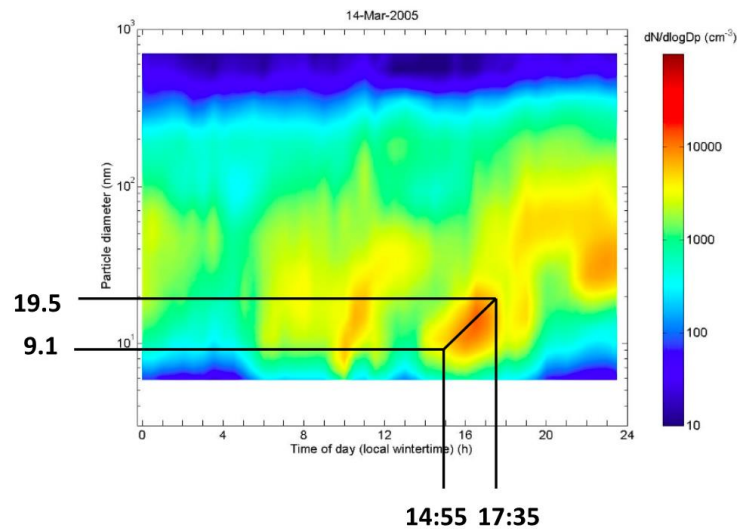


Figure 4. Image showing a linear approximation of the growth rate of a nucleation event in Lille Valby, Denmark on March 14, 2005.

5.3.5 Lognormal Fitting of Size Distribution Data

In order to determine the number of particles that could be attributed to new particle formation from Lille Valby, the size distribution data from Vavihill was analyzed using a multi lognormal fitting algorithm (Hussien et al. 2005). This algorithm parameterizes aerosol particle number size distributions using the multi lognormal distribution function. It was used to obtain log-normal parameters such as the mode number concentration, geometric variance and geometric mean diameter. The model used consists of up to three lognormal modes in the fine particle size range. For a more detailed breakdown of the algorithm see Hussien et al. 2005.

For this analysis the diameter range of the particle population in question was determined and then used to relate that population to its corresponding mode and number concentration. Therefore the number of particles which travelled from Lille Valby to Vavihill was quantified.

5.3.6 Reclassification

In this stage of analysis each event day was visually inspected. Wind speed, growth rate, and particle size from the lognormal fitting was used. First, a visible connection had to be established. In some cases, there was no clear group of particles in Vavihill that could be traced to Lille Valby. It was then determined whether this connection was plausible using the wind speed and time separation of the particles. For example, if particles began at time 12:00 in Lille Valby and began at time 14:00 in Vavihill with a travel time of 2 hours the connection was approved. The final step was to check that the size of particles in Vavihill could be connected back to Lille Valby using the particle size from the lognormal fitting and the previously calculated growth rate. The total number of particles and the number of particles belonging to the mode that could be traced back to Lille Valby was recorded at six hour intervals starting at 0:00. The size at additional hours was recorded when necessary. Below, Figure 3 shows an example of an approved date. The left image is the data from Lille Valby and the right image is the data from Vavihill. The event in question is circled in black for clarity.

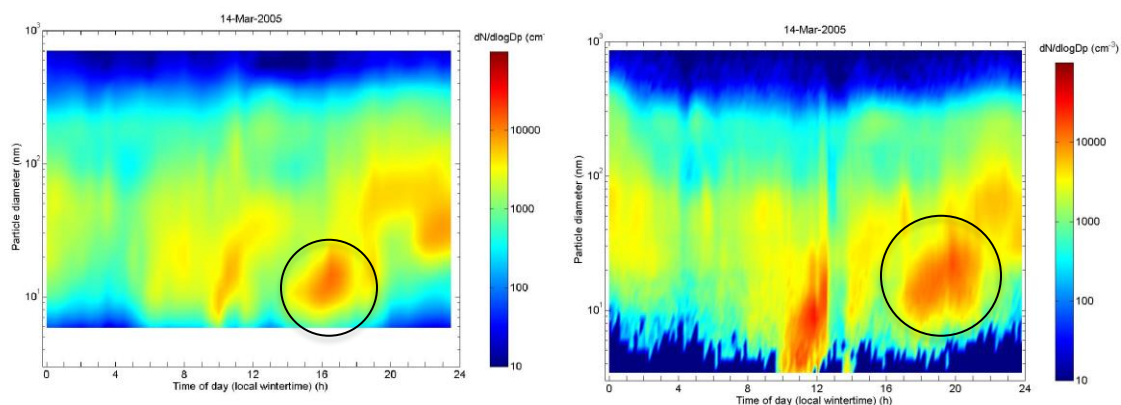


Figure 5. Reclassified new particle formation event in Vavihill (right) from Lille Valby (left).

6. Results

6.1 General Vavihill Results

Prior to reanalysis, results from the Vavihill station showed that 10.4% of 1825 days were classified as new particle formation events. Of the 1825, 1133 were classified as either undefined, non-event or unclear event days. From this 1133, 44 days corresponded with a type 1 event in Lille Valby and were selected for analysis.

6.2 Reanalysis of NPF

After analyzing all of the selected cases it was found that 44 out of 1480 possible days in Vavihill corresponding to a type 1 event in Lille Valby could be reclassified as new particle formation events. This percentage represents days where the wind trajectory showed particles travelled from Lille Valby to Vavihill and grew at the same rate.

Of the cases analyzed, 27 were removed due to wind trajectories showing that the air mass in Vavihill could not have come from Lille Valby. This included any cases where the wind came from the north, south or east of Vavihill. Cases where wind travelled over open sea were also excluded with the assumption that the particles would then have been affected by marine aerosols as well as ship traffic. Due to a lack of particles found at the Lille Valby station, 7 more events were removed. These instances made it difficult to calculate the growth rate of the particles and determine a clear connection between particles at the two sites. Furthermore 2 events were removed as it was found that the growth rates from the two locations did not match. A single event was removed due missing data in Vavihill. This left 7 days to be reanalyzed for NPF from Lille Valby.

The average percentage of particles that can be attributed to new particle formation from Lille Valby on reclassified days over the five year period considered is 64.18%. When the days eligible for reclassification are considered, particle numbers are taken from the lognormal fitting data at 6 hour intervals (perhaps more frequently where events did not adequately fit these intervals). Only the times where an event was occurring were considered (the percentage for all other times is 0). The results are shown below in Table 2.

Table 2. *Reclassified Particles*

Event Type in VVHL	Date (YY-MM-DD)	Time	Number of Particles Reclassified	Total Number of Particles	% Reclassified
2	05-03-14	18:00	4235.608	5980.082	70.83
		24:00:00	3060.254	3972.583	77.03
2	05-09-13	18:00	1788.634	4677.44	38.24
2	06-05-15	6:00	2999.241	3887.464	77.15
		18:00	5069.812	5533.539	91.62
		24:00:00	949.422	3070.484	30.92
-1	06-08-09	18:00	3531.945	5385.621	65.58
		24:00:00	4062.482	4201.091	96.70
	06-08-10	6:00:00	1923.817	2334.183	82.42
2	09-10-02	22:00	2700.718	3151.105	85.71
		24:00:00	3485.953	4400.177	79.22
	09-10-03	6:00	2012.448	2757.301	72.99
-1	10-06-19	24:00:00	181.921	855.988	21.25
2	10-07-31	12:00	1636.165	4870.277	33.59
		18:00	2043.424	5190.565	39.37
Average					64.18

7. Discussion

The reanalysis method found that 64.18% of the particles analyzed in Vavihill, Sweden could be reclassified as originating from new particle formation in Lille Valby, Denmark. This result applies to the days where wind trajectories and growth rates matched for the two stations. Although only 7 cases were found to match for this specific wind direction, the implications of this result are still found to be significant. If this result was extrapolated for other wind directions, the amount of particles reclassified would increase substantially. Therefore the amount of newly formed particles which can grow to be condensation nuclei in this area would seemingly increase.

As knowledge of aerosol particles continues to grow, so does their perceived effect on the Earth's climate. Prior to reclassifying these particles their origin was unknown, therefore their ability to affect climate was also unknown. Now knowing how much more of these particles are related to new particle formation their effect on climate can be quantified and accounted for. This climate effect might be estimated with the method defined by Kurten et al. (2003) mentioned in section 3.5.

Knowing that these particles come from NPF and can grow to sizes big enough to form cloud condensation nuclei affects what is known about climate in southern Scandinavia and the world. On a global scale, this reanalysis may help to formulate an assumption about the effect aerosol particles have on radiative forcing. The effect that is currently believed to exist may very well be much lower than the actual effect felt by the Earth's atmosphere. For instance, the degree to which greenhouse gas emissions have been affecting climate could be much higher than previously thought due to the cooling effect of aerosols. Overall these results contribute to the global aerosol model increasing the degree of this effect and how much is known about it.

As previously mentioned, it would be beneficial to extend the analysis performed in this study by determining the percentage of unclear event and nonevent days that were able to be reclassified for this specific wind direction. To do this all 890 of these days in Vavihill would need to be analyzed for wind trajectory. By finding the number of days where aerosol particles could have travelled from Lille Valby to Vavihill, a percentage for the ability to classify this type of day could be determined for that wind direction. For example, assume that 50 of all unclear event days in Vavihill were found to have wind patterns coming from Lille Valby. It could then be said that 5/50 or 10% of all unclear event days could be reclassified as new particle formation days for this wind direction. This result could then be extrapolated for all wind directions reaching Vavihill. As shown by this study, 0% of undefined days could be reclassified for any wind direction using this method.

Buenrostro et al. (2005) was able to reclassify 90% of 1630 previously undefined days of data collected at Hyytiälä, Finland as failed events, pollution related peaks, or ultrafine mode concentration peaks using the method described in section 4.3. It is possible that by combining the application of the Buenrostro method as well as the new method developed in this study, the fraction of days left unclassified would be even smaller.

Hussein et al. (2009) found that, by reviewing the time span and spatial scale of regional new particle formation events in Finland and southern Sweden at several different stations, the events could be traced to distance of over 220 km upwind of where they were observed. When assessing each day for reclassification it was noticed that some days had similar growth shapes in Lille Valby and Vavihill but began at the same time. See Figure 4 below for an example. It is possible that this could be new particle formation from another region travelling over a large distance reaching Lille Valby and Vavihill at the same time. Due to the limited number of stations globally to analyze the aerosol particle concentrations it is difficult to predict the origins of these particles. However, if particles formed outside of the Scandinavian region, farther than Lille Valby, are travelling and affecting the area around Vavihill, the effect of new particle formation could be much greater than it is currently believed to be.

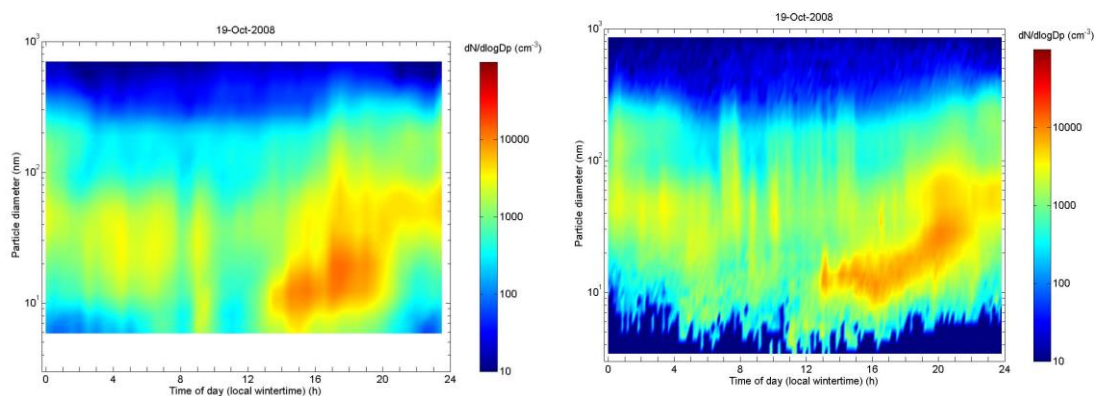


Figure 5. New particle formation event in Lille Valby (left) and Vavihill (right) both starting at 13:00.

8. Conclusions

The goal of this study is to better understand atmospheric aerosol particle nucleation and how aerosol particles travel through the atmosphere. A new method to reclassify particles at a station in Vavihill, Sweden was proposed by using new particle formation event data from another station in Lille Valby, Denmark. The particles considered for reclassification included those from undefined, non-event and unclear event days in Vavihill. Over half of the particles (64.18%) in Vavihill during reclassified events are now accounted for by this new method. With this knowledge, more analyses, such as the ones discussed above, could be performed to determine the effect of these particles travelling in the atmosphere. By extrapolating the data obtained a bigger effect can be seen and therefore quantified to determine the true effect aerosol particles have on global radiative forcing.

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