Investigation of noctilucent cloud properties and their connection with solar activity

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« For example: the Mexican sierra has "XVII-15-IX" spines in the dorsal fin. These can easily be counted. But if the sierra strikes hard on the line so that our hands are burned, if the fish sounds and nearly escapes and finally comes in over the rail, his colors pulsing and his tail beating the air, a whole new relational externality has come into being - an entity which is more than the sum of the fish plus the fisherman. The only way to count the spines of the sierra unaffected by this second relational reality is to sit in a laboratory, open an evil-smelling jar, remove a stiff colorless fish from formalin solution, count the spines, and write the truth "D. XVII-15-IX." There you have recorded a reality which cannot be assailed – probably the least important reality concerning either the fish or yourself. »

*John Steinbeck*
Noctilucent clouds (NLC) are optically thin layers of water ice particles present near the summer mesopause at high latitudes. Because of their extraordinary height of about 83 km, they can become visible to the naked eye when the sun sinks between 6 and 16° below the horizon, providing a dazzling display of bluish light.

The observation of NLC conveys unique information concerning the different processes taking place in the upper mesosphere. Since NLC are extremely sensitive to changes in temperature and water vapor content, it is possible, by studying the variation of NLC properties on different timescales, to gain insights into the various atmospheric processes in action near the summer mesopause. Understanding which physical mechanisms are important for this region of the atmosphere is also crucial for quantifying the possible change in NLC properties which could be attributed to secular trends as a result of anthropogenic activity.

The purpose of this thesis is to analyze NLC properties measured from the space-borne SCanning Imaging Absorption spectroMeter for Atmospheric ChartograpHY (SCIAMACHY) instrument. Limb observations of NLC scattered sunlight, in the spectral range 264-300 nm, are exploited for the retrieval of NLC occurrence, radiance, and altitude. The good spectral resolution of the instrument also permits the determination of the NLC particle size, provided assumptions are made on the particle shape and size distribution. The modeling of the particle scattering is based on the T-matrix approach. The NLC particle size can only be unambiguously retrieved in the northern hemisphere due to the particular geometry of observation.

A climatology of NLC occurrence frequency, radiance, altitude, and particle size is presented for years 2002-2009 and for both hemispheres. The seasonal variations of NLC properties are in good agreement with independent data sets. It is also shown that despite the coarse vertical resolution of SCIAMACHY, the daily NLC altitudes correlate well with the supersaturation levels, especially in the northern hemisphere. SCIAMACHY’s global coverage grants the possibility of observing the response of NLC activity linked to various atmospheric phenomena such as the solar proton events, planetary waves, and the zonal dependence of gravity wave forcing. The dependence of the NLC particle size on latitude, local time, and altitude is also investigated. It is shown that the NLC particle radii increase with latitude, decrease with increasing altitude and are slightly larger for a local time of 9 PM compared with 11 AM. A study of the sensitivity of the retrieved NLC particle size
on the assumptions is also presented, showing a large variation of the NLC particle size depending on the PSD width and the particle shape. The SCIAMACHY retrieved NLC particle radii are also compared with different independent particle size data sets, revealing how much information can be gained by merging different data sets together.

Finally, the effect of the 27-day solar cycle on NLC properties is investigated. An analysis based on the SCIAMACHY and the Solar Backscattering UltraViolet (SBUV) data sets shows that there is a clear anti-correlation of NLC occurrence rate and albedo with solar activity on short timescale, while the NLC daily altitude is positively correlated with solar activity. The response of the NLC properties to the 27-day solar forcing is generally larger at higher latitudes. The results of the analysis with the SBUV data set reveal that the sensitivity of the NLC albedo to the 27-day solar cycle is similar to that of the 11-year solar cycle, suggesting that similar mechanisms may be responsible for the variation of NLC properties. Inspection of the temperature and water vapor from the Microwave Limb Sounder (MLS) indicates that temperature, not water vapor, is responsible for the variation in NLC properties.
Publications

Journal Articles


Conference Contributions


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 introductory text : "I have a friend who’s an artist, and he sometimes takes a view which I don’t agree with. He’ll hold up a flower and say, "Look how beautiful it is," and I’ll agree. But then he’ll say, "I, as an artist, can see how beautiful a flower is. But you, as a scientist, take it all apart and it becomes dull." I think he’s kind of nutty. First of all, the beauty that he sees is available to other people and to me, too, I believe, although I might not be quite as refined aesthetically as he is; but I can appreciate the beauty of a flower. At the same time, I can see much more about the flower than he sees. I can imagine the cells in there, the complicated actions inside which also have a beauty. I mean it’s not just beauty at this dimension of one centimeter, there is also beauty at a smaller dimension, the inner structure. Also the processes, the fact that the colors in the flower evolved in order to attract insects to pollinate it is interesting - it means that insects can see the color. It adds a question: Does this aesthetic sense also exist in the lower forms? Why it is aesthetic? All kinds of interesting questions which shows that a science knowledge only adds to the mystery and awe of a flower. It only adds. I don’t understand how it subtracts. »

Richard Feynman

1

Introduction

1.1 Motivation and goals

Noctilucent clouds (NLC), also referred to as polar mesospheric clouds (PMC), are optically thin layers of water ice particles (Hervig et al., 2001) present near the summer mesopause at high latitudes. These clouds are optically too thin to be witnessed for a ground observer by daylight but because of their extraordinary height of about 83 km, they can become visible to the naked eye when the sun sinks between 6 and 16° below the horizon (Gadsden and Schroeder, 1989), providing a dazzling display of bluish light (see figure 1.1). Hence, the name noctilucent (or night shining) clouds.

The formation of clouds in such a remote and dry region of the atmosphere puzzled researchers for some time. One perplexing aspect of NLC is that the first sighting of these mesospheric clouds has been reported in 1885 by Backhouse (1885), begging the question whether these clouds existed before. It has been argued that the Krakatoa eruption in 1883 could have been responsible for abundant water vapor injection in the upper atmosphere and would have led to the formation of NLC at lower latitudes, explaining the many independent sightings in Germany and Russia in 1885. The Krakatoa eruption might also have had an indirect effect on the sighting of NLC since the particulate matter injected in the atmosphere gave rise to stunning sunsets, thereby increasing the number of potential observers of NLC.

Another possible reason for the late appearance of NLC is related to the increase in greenhouse gases concentration in the atmosphere since the beginning of the industrial revolution, presumed to have had a cooling effect in the upper layers of the atmosphere
(Roble and Dickinson, 1989), which would improve the condition of NLC formation. The doubling of atmospheric methane since the last century (Khalil and Rasmussen, 1994) is believed to have produced a 50% increase in mesospheric water vapor content, also making the formation of NLC more likely. For these reasons, NLC have been suggested as possible early indicators of global change, since the upper layers of the atmosphere should be far more sensitive to changes in greenhouse gases increase than near the surface (Thomas, 1996). This effect would manifest itself in the form of brighter NLC extending to lower latitudes. However, there is considerable debate about the actual presence of changes in the mesosphere (Lübken, 2000) or whether NLC would actually be a good indicator of atmospheric changes (von Zahn, 2003).

Not only are NLC possible indicators of global climate change, but they are also unique in the information they convey about the different processes taking place in the upper mesosphere. The mesosphere has long been coined the “ignorosphere” due to the lack of data concerning this remote region of the earth, where only rocket flights can provide in-situ measurements. Since NLC are extremely sensitive to changes in temperature and water vapor content, it is possible, by studying the variation of NLC properties on different timescales, to gain insights into the various atmospheric processes in action near the summer mesopause. Understanding which physical mechanisms are important for this region of the atmosphere is also crucial for quantifying the possible change in NLC properties which could be attributed to secular trends as a result of anthropogenic activity.

The goal of this project is to use SCIAMACHY’s measurements to retrieve relevant information relative to noctilucent clouds. Other than common macroscopic properties of NLC such as the occurrence rate, radiance and altitude, the excellent spectral resolution of SCIAMACHY and the good signal-to-noise ratio of NLC limb measurements permits the retrieval of microscopic properties of NLC, namely their particle size. These retrieved NLC properties are will be surveyed and analyzed in light of the present understanding of NLC and see if they are in accordance with results obtained through different experimental means.
A further goal of this thesis is to use NLC properties from SCIAMACHY, as well as various other data sets, in order to understand important processes taking place at the summer mesopause linked with solar variability on a 27-day timescale. There is already evidence concerning the interaction of the sun with the atmosphere, but very few studies regarding how the mesosphere is influenced by solar activity on a the timescale of a solar rotation. One aspect of this work is to investigate whether solar variability on short timescale affects NLC properties and, if it is indeed the case, how exactly this interaction takes place.

1.2 Structure of this thesis

This thesis is structured as follows:

Chapter 2 introduces fundamental concepts relevant for this work, including a description of the sun as a variable star as well as covering some aspects of light-matter interaction taking place in the atmosphere, especially scattering.

Chapter 3 describes in some detail the structure and composition of the earth's atmosphere, with an emphasis on the summer mesopause region near the pole. The physical processes responsible for making the polar summer mesopause the coldest place on earth are reviewed and some characteristics of noctilucent clouds formation and their interaction with the atmosphere are also presented.

In Chapter 4, the instruments and data sets used throughout this work are presented. First and foremost, the characteristics of the SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY) are surveyed, as NLC properties will be derived from SCIAMACHY’s measurements. The chapter also covers the important features of the various data sets exploited repeatedly throughout this work, as well as the uncertainties associated with them.

Chapter 5 details the algorithm used to retrieve NLC occurrence frequency, radiance, altitudes and particle size with SCIAMACHY. Errors on the retrieved parameters are also estimated.

Chapter 6 presents the results of a climatology of NLC occurrence frequency, radiance and altitude retrieved since August 2002 with SCIAMACHY. Physical mechanisms responsible for intra-seasonal variation of NLC properties are discussed and comparisons with different data sets are also presented.

Chapter 7 focuses on the analysis of the retrieved NLC particle size. The spatial, seasonal and year-to-year variations will be described, along with an analysis of the connection between the NLC particle radii and the latitude, altitude and local time of observation. SCIAMACHY’S retrieved NLC particle size is also compared with independent data sets.
In chapter 8, the effect of the 27 day solar activity on NLC properties is investigated using the SCIAMACHY and SBUV data sets and different techniques, including the so-called superposed epoch analysis. The sensitivity of the NLC response to the 27 day solar cycle is presented, and the associated sensitivities in temperature and water vapor content are also described.

Chapter 9 shall summarize the major findings of this work.
This chapter introduces fundamental concepts of physics relevant for this work, encompassing the sun as a dynamic source of energy for the earth and the interaction of matter and radiation in the atmosphere. These concepts are of vital importance, first and foremost to the basic understanding of the atmosphere, but also to appreciate specific aspects of this work, such as the interaction between solar variability and the summer mesopause as well as the retrieval of NLC particle size from spectral measurements of scattered sunlight.

2.1 The sun

The energy that the sun provides to our planet is at the root of the lively and dynamic state of the atmosphere. Figure 2.1a pictures the structure of the sun as it is presently understood. This enormous gaseous orb, $6.96 \times 10^5$ km in radius and with a mass of $1.99 \times 10^{30}$ kg, produces $3.83 \times 10^{26}$ W of power through fusion reactions at the core of the sun, involving mostly the conversion of four hydrogen nuclei into one helium nucleus and energy leading to temperatures of the order of $10^8$ K (Lewis, 2004).

It was estimated by Mitalas and Sills (1992) that it takes an average time of about $10^5$ years for a photon to diffuse from the solar core to the surface. Throughout this long journey, photons are absorbed by the plasma and remitted many times in all directions until they reach the zone of convection. From here, energy is transmitted by means of electromagnetic radiation and by the convective motion of the gas driven by the steep temperature gradient between the hot radiative zone ($\approx 10^6$ K) and the relatively cold...
surface of the sun (≈ 6000 K), also called the photosphere. From the photosphere, radiation is finally emitted to space in the form of a continuous electromagnetic spectrum. Because cooler gases in the upper photosphere and in the chromosphere will absorb some of this radiation, the solar spectrum seen from space contains absorption lines. These are the so-called Fraunhofer lines, named after the German scientist Joseph von Fraunhofer who recorded 574 lines of the solar absorption spectrum. Figure 2.1b shows a vertical temperature profile of the solar atmosphere as well as the origin of different absorption lines within this region of the sun (Liou, 2002).

In general, the photosphere can be modeled as following Planck’s law of the blackbody which states that the spectral distribution of the electromagnetic blackbody emission should be only dependent upon the temperature of the blackbody:

\[ B_\lambda = \frac{2\pi h c^2}{\lambda^5} \left( e^{\frac{hc}{\lambda kT}} - 1 \right)^{-1} \]  

(2.1)

where \( B_\lambda \) is the energy emitted per unit surface in an infinitesimal wavelength interval d\( \lambda \), \( h \) is Planck’s constant, \( c \) is the speed of light, \( k \) is Boltzmann’s constant, \( \lambda \) is the wavelength and \( T \) is the blackbody temperature. Figure 2.2a shows SCIAMACHY irradiance measurements of the solar spectrum at the top of the earth’s atmosphere (TOA) along with the modeled solar irradiance at incident on earth (dashed line). The latter is calculated from Planck’s law assuming the sun is a blackbody with a temperature of 5800K and scaling it by a geometrical factor according to:

\[ S_\lambda = \pi B_\lambda^{5800} \left( \frac{R_E}{D_{ES}} \right)^2 \]  

(2.2)

where \( R_E \) is the average earth’s radius (6.371 × 10^6 m) and \( D_{ES} \) is the average earth-sun distance (1.5 × 10^{11} m). The measured and modeled irradiances are generally in good agreement.
agreement, although one can see some deviation from the blackbody model. Figure 2.2b shows such deviations in more detail for the ultraviolet (UV) portion of the solar spectrum, alongside blackbody spectral irradiance curves assuming different blackbody temperatures.

Besides electromagnetic radiation, particles are also emitted from the corona, mainly in the form of protons and electrons, to a lesser extent He$^{2+}$ ions and only traces of heavier ions. Due to the very high coronal temperatures and the wave activity which can transport energy from the bottom of the photosphere up to the corona, particles there tend to have large kinetic energies. Particles can therefore be energetic enough to overcome the gravitational potential of the sun, resulting in a stream of particles radiating from the corona through the solar system: this is the so-called solar wind.

Although the sun might appear as being unchanging, it is, on the contrary, quite a dynamic celestial body. The primary cause for the variation of solar activity is due to changes in magnetic fields generated by a self-exciting magnetohydrodynamic dynamo in the sun’s interior, which in turn affects the properties of the solar atmosphere and therefore the electromagnetic and corpuscular output (Tsiropoula, 2003). The second cause of the apparent modulation of solar activity as seen from the earth is a consequence of the solar rotation, westward in direction with an angular speed dependent on the latitude. The mean synodic period of rotation at the equator is 26.24 days and increases to 35 days at the poles (Moussasa et al., 2005). The rotation modifies the coverage of solar active regions facing the earth which also affects the incoming electromagnetic radiation and particle stream. Since the average modulation of solar parameters is usually close to a 27-day period, it is often referred to as the 27-day solar cycle.

Solar activity can change over very different timescales, from minutes (i.e. solar flares) to billions of years through the stellar evolution. The most apparent periodic modulation of solar activity is the 11-year cycle, which is actually a manifestation of the 22-year (Hale)
Lyman-α irradiance [10\(^{11}\) photons cm\(^{-2}\) s\(^{-1}\)] anomalies B A C anomalies

Figure 2.3: Composite solar Lyman-α irradiance time series (Woods et al., 2000). Panel A: 11-year cycle in the Lyman-α irradiance. Panel B: Lyman-α irradiance time series with the 11-year component removed. One can see that the 27-day cycle is modulated by the 11-year cycle. Panel C: Details of the structure of the 27-day variation for year 1989 in the Lyman-α anomaly time series.

cycle if we take into account the polarity of the magnetic dipole (Kane, 2002). Indeed, every 11 years the magnetic dipole changes polarity, and hence only every 22 years the magnetic configuration of the sun is back in its original state. The observation of the cycle was first carried out through sunspot examination. Sunspots are colder regions at the photospheric level created through the effect of strong magnetic field lines in the convective zone hindering the convective motion of the gas and hence the transfer of heat. This effect cools parts of the photosphere to temperatures of about 4000 K which, due to their lower temperatures, appear darker. Sunspot dimensions are on the order of several thousands of kilometers and can exist for a few days if small, but also several weeks if large enough. They usually tend to group in active regions, the surrounding of which is brighter and is called a faculae. It might appear paradoxical that an increasing number of sunspots at the sun’s surface lead to a larger total solar irradiance (TSI) incident on the earth. However, this phenomenon can be explained by the fact that the darker sunspots are counterbalanced and even outweighed by an increase in irradiance from the brighter faculae and the presence of the active network. Overall, the TSI changes by about 0.1% over the course of the 11-year cycle, corresponding to an absolute variation of 1.3 W m\(^{-2}\). The irradiance at different wavelengths however can change by up to a few percents for wavelengths below 300 nm (Pagaran et al., 2009).

During periods of high solar activity, there will also be more occurrences of solar flares and coronal mass ejections (CME), which can lead to solar proton events, a sporadic flux of high-energy protons and other energetic particles. These protons can sometimes have enough energy to penetrate the earth’s magnetic field and reach the atmosphere, where they can have a substantial effect on composition and temperature (Jackman et al., 1999; López-Puertas et al., 2005). The solar activity also has an effect on the interplanetary magnetic field (IMF), carried along by the solar wind. The stronger field generated during periods of high activity deflects more efficiently galactic cosmic rays and will therefore modulate the input of these cosmic rays in the earth’s atmosphere and have an effect on ion production and spallation reactions in the upper atmosphere (de Wit and Watermann, 2009).
Many different proxies of solar activity exist. The most commonly found in literature are the solar radio flux emitted at 10.7 cm, the core-to-wing ratio of the Mg II line at 279.9 nm, the intensity of the bright H Lyman-\(\alpha\) line at 121.57 nm and the TSI measured above the atmosphere. Since these proxies reflect the level of solar activity, they also exhibit periodic features associated with the sun such as the 11-year and 27-day cycle. Figure 2.3 shows an example of such a proxy, the Lyman-\(\alpha\) irradiance, between 1975 and 2009. The 11-year cycle is very prominent if figure 2.3a. The removal of the 11-year component (figure 2.3b) emphasizes the 27-day solar rotation period, which is also modulated in amplitude by the 11-year cycle. A close-up of panel B is presented for year 1989 in Figure 2.3c where the variability of the 27-day cycle is shown in more detail. A study of this proxy shows that the mean amplitude of the 27-day cycle at 121.6 nm corresponds to about 25% that of the 11-year solar cycle. This time series will be used extensively in chapter 8 to study the effect of the 27-day variation in solar activity on NLC properties. The method used to assemble this composite time series will be described in section 4.2.

2.2 Basic radiative transfer in the atmosphere

The sun is the main source of energy for the earth’s atmosphere. Its radiation incident on the earth interacts in different ways with the atmospheric constituents. This section will cover the most important processes by which radiation and atmosphere interact.

Absorption

One of the most important forms of interaction between radiation and matter is absorption, in which energy from the electromagnetic field is transferred to the physical medium. The absorption of radiation is mathematically modeled by the Beer-Lambert law which states that the absorbed intensity \(dI(\lambda)\) along the infinitesimally small optical path \(ds\) in a medium is directly proportional to the incoming radiation intensity \(I(\lambda)\), the absorption cross-section \(\sigma_a\) (per particle) and the number density \(N(s)\):\n
\[
dI(\lambda) = -I(\lambda) \sigma_a N(s) ds\tag{2.3}
\]

The minus sign on the right hand side denotes the loss of intensity from the incident beam to the medium. Integrating eq. 2.3 over an arbitrary optical path \(s\), we find

\[
I(\lambda) = I_o(\lambda) e^{-\int \sigma_a N(s) ds}\tag{2.4}
\]

where \(I_o(\lambda)\) is the incident intensity at \(s = 0\). Eq. 2.4 is also sometimes written as:

\[
I(\lambda) = I_o(\lambda) \cdot e^{-\int k_\lambda(s) ds} = I_o(\lambda) \cdot e^{-\tau(\lambda,s)}\tag{2.5}
\]
where $k_a(\lambda,s)$ is the absorption coefficient and $\tau(\lambda,s)$ is the optical depth, also referred to as optical thickness. The absorption cross-section is dependent on the constituents and is a strong function of wavelength. In the atmosphere, light is absorbed by atoms, molecules and aerosols. Atoms absorbing radiation can be excited, undergoing electronic transitions depending on their electronic configuration. Such transitions require radiation with energy in the range of a few eV, i.e. with a wavelength typically in the UV-visible range, while ionization of an atom is achieved with X-ray and extreme ultraviolet (EUV) photons. Molecules can be subjected to ionization, photodissociation as well as electronic, vibrational and rotational transitions. Excitation of a molecule in different vibrational and rotational states can be initiated by radiation with wavelengths in the infrared (IR) and in the far IR/microwave range respectively.

The penetration of radiation in the atmosphere will depend strongly on the absorption cross-section of all the atmospheric species combined, as well as their respective abundance. Figure 2.4 shows the penetration depth of solar radiation with $\lambda < 310$ nm in the earth’s atmosphere, defined as a reduction by a $1/e$ factor in intensity. The figure also indicated the species mainly responsible for absorption in a given wavelength range and the convention for naming the different parts of the electro-magnetic (EM) spectrum in the lower part of the graph. One sees that the UV-B radiation, responsible for skin erythema, is generally not completely absorbed above the surface depending on the O$_3$ optical depth. Another interesting point is the fact that the strong Lyman-α emission line from the sun is situated in a minimum of the O$_2$ absorption cross-section, which means that it can penetrate down to about 70 km.

The absorption of solar radiation has thermal and photochemical consequences on the state of the atmosphere. Radiation absorbed by O$_3$ and O$_2$ in the upper and middle atmo-
The presence in the atmosphere of small particles, atoms and molecules also affects the electromagnetic field of the incoming radiation by redistributing it spatially through scattering processes as illustrated in figure 2.5. Two categories of scattering exist: elastic and inelastic. Elastic scattering implies a change in the direction of propagation of the incoming radiation but without a loss in energy, so that elastically scattered radiation has the same wavelength as that of the incident radiation. The most common types of scattering which occur in the earth’s atmosphere, Rayleigh and Mie scattering, are of this type. Inelastic scattering involves both a redistribution of the direction of propagation and a gain (or a loss) of energy of the incident radiation. Raman scattering is an example of such a process, where scattered photons have a larger wavelength than the incident ones. Raman scattering is responsible for the so-called Ring effect, filling in some of the Fraunhofer absorption lines (Chance and Spurr, 1997). As Raman scattering is not relevant for the present study, only Rayleigh and Mie scattering will be reviewed here.

One way of defining scattering processes is through the concept of the phase function \( P(\theta) \) which describes the angular distribution of the scattered energy. It is defined as:

\[
P(\theta) = \frac{4\pi}{\sigma_{\text{scat}}} \frac{dC_{\text{scat}}}{d\Omega}
\]  

(2.6)
where $\sigma_{\text{scat}}$ is the scattering cross-section per molecule and $\frac{dC_{\text{scat}}}{d\Omega}$ is the differential scattering cross-section, which describes the amount of radiation scattered at a given angle $\theta$ and within a given solid angle by a molecule. The scattering angle $\theta$ is defined as the angle between the direction of the propagation of the incident radiation and that of the scattered radiation (figure 2.5). The difference between the phase function and the differential scattering cross-section is that the phase function is normalized according to the following condition:

$$\int_{\Omega} P(\theta) \frac{4\pi}{d\Omega} = \int_{0}^{2\pi} \int_{0}^{\pi} P(\theta) \frac{4\pi}{\sin \theta} d\theta d\phi = 1 \quad (2.7)$$

The scattering cross-section $\sigma_{\text{scat}}$ is analogous to the absorption cross-section $\sigma_a$ since it describes the amount of radiation subtracted from the incident beam in the direction of propagation. Both the scattering cross-section and the differential scattering cross-section are generally dependent on the wavelength of the radiation, the shape and the size of the scatterers.

**Rayleigh scattering**

Assuming that we have spherical particles with radii $r$ as scattering centers, we can define the dimensionless size parameter $a$:

$$a = \frac{2\pi r}{\lambda} \quad (2.8)$$

One notices that $a$ is equivalent to the ratio of the circumference of the particle and the wavelength. For the instance where $a << 1$, i.e. where the size of the scatterer is very small compared to the wavelength, Rayleigh scattering takes place. This is the case for the interaction of solar radiation and atmospheric gases since molecular sizes are of the order of $10^{-10}$ m. Although molecules are not spherical as was assumed in the derivation of the solution by Lord Rayleigh (Rayleigh, 1871), the exact shape of the scattering center is usually of little importance due to the small size of the particles relative to the wavelength and can, most of the time, be treated as a sphere of equivalent volume. Another assumption made by Rayleigh when solving the problem of scattering by small particles is independent scattering. This ensures that the particles are sufficiently far from each other so that no cooperative effect takes place, such as is the case in crystals for instance.

The phase function of the Rayleigh scattering for unpolarized light (e.g., solar radiation) is given by

$$p_{\text{Ray}}(\theta) = \frac{3}{4} \left( 1 + \cos^2(\theta) \right) \quad (2.9)$$

The Rayleigh scattering cross-section is computed as follows

$$\sigma_{\text{Ray}} = \frac{128 \pi^5 \alpha^2}{3 \lambda^4} \quad (2.10)$$
with the polarizability $\alpha$ defined as

$$\alpha = \frac{3}{4\pi N} \left( \frac{m^2 - 1}{m^2 + 2} \right)$$

Figure 2.6: Panel A: Rayleigh phase function Panel B: Mie phase function for ice particles of different sizes and incident radiation with $\lambda = 290$ nm.

Mie Scattering

Mie scattering is the far-field solution for an electromagnetic plane wave scattered by a dielectric sphere, regardless of the size parameter. It is usually used to explain the interaction between solar radiation and aerosols. The solution was worked out independently by many different researchers (Clebsch, 1863; Lorenz, 1890; Debye, 1909; Mie, 1908) but bears the name of the German physicist Gustav Mie who solved the problem and published its solution in 1908 to explain laboratory measurements made by one of his students.
Figure 2.7: Mie scattering cross-section for ice particles of different sizes and incident radiation with $\lambda = 265$ nm. Panel A shows the functional relationship on a logarithmic scale while panel B presents the results on a linear scale, emphasizing the Mie resonance for large particle size.

Since the mathematical apparatus to calculate Mie scattered radiation properties is substantial, no details of the equations are given here but they can be found in a number of publications (e.g. van de Hulst, 1957; Bohren and Huffman, 1983). The difference between Rayleigh scattering and Mie scattering, in terms of phase function, is illustrated in figure 2.6b, for $\lambda = 290$ nm incident radiation on particles of different sizes. The Rayleigh phase function is represented by the grey filled area for reference purposes, and one notices that for a small (10 nm) particle, the Mie phase function is essentially the same as that calculated by the Rayleigh scattering model. As the size of the particle (or more generally, the size parameter) increases, however, the phase function becomes more asymmetric, favoring forward scattering over backscattering. An important property of Mie scattering is that the cross-section $\sigma_{\text{Mie}}$ is a strong function of the particle size, proportional to $r^\chi$ over a limited range of the size parameter. The exponent $\chi$ is usually between 4 and 6 for size parameters $\alpha < 1$. Figure 2.7 shows the cross-section for scatterers of different radii with an incident radiation of 265 nm. The log-log plot from panel A highlights the rather constant value of $\chi = 6$ for smaller particle sizes, which quickly changes as $\alpha \approx 1$, and after that point, enters in a resonance regime which is more clearly depicted in the linearly scaled figure 2.7b.

One limitation of the Mie solution to the scattering problem is that it assumes spherical particles. It is however possible to solve the problem for a certain number of geometries (e.g., spheroids, cylinders) efficiently using different approaches such as the discrete dipole approximation (Hage et al., 1991) or the T-matrix formulation (Waterman, 1965). The effect of the shape on the phase function is shown in figure 2.8 for spheres, prolate and oblate spheroids with axial ratio (AR) of 0.2 and 5.0 respectively. As can be seen from figure 2.9, prolate spheroids are elongated spheres which can be used to describe needle-like particles while oblate spheroids are compressed spheres, similar to the actual shape of the earth. A spheroid particle volume has a tendency to scatter more light in the forward direction than spheres, for a volume equivalent radius.
2.2 Basic Radiative Transfer in the Atmosphere

Figure 2.8: Scattering phase function for ice particles of different sizes, incident radiation with $\lambda = 290$ nm and shape. Panel A: phase function of spherical particles. Panel B: phase function of prolate spheroids with axial ratio 0.2. Panel C: phase function for oblate spheroids with axial ratio 5.0.

Figure 2.9: A schematic of oblate and prolate spheroids.

For Rayleigh scattering, the amount of radiation scattered is proportional to the number of scattering centers, independently of their size as long as the condition $a << 1$ is fulfilled. On the other hand, the effective Mie scattering of an ensemble of particles with normalized size distribution $f(r)$ is highly influenced by the actual shape of the distribution. The effective differential scattering cross-section for an ensemble of particles, calculated according to the assumption of independent scattering, is

$$\frac{d\sigma^*_{\text{scat}}}{d\Omega} = N \int_{0}^{+\infty} \frac{d\sigma_{\text{scat}}(\lambda, r, \theta)}{d\Omega} \cdot f(r) \cdot dr$$

(2.12)

where $N$ is the particle density and the star ($^*$) denotes an effective property of the ensemble of particles. There exist many different particle size distributions (PSD) which can model an ensemble of atmospheric aerosols. The PSD depends on the formation processes of aerosols.
Figure 2.10: Normal and lognormal distribution of particle sizes. Also shown is the contribution of different parts of the population to the effective scattering cross-section for $\lambda = 265$ nm.

and their interaction with their environment. As will be shown later, two PSDs play an important role in the modeling of NLC particles: the normal and the lognormal distribution, described by the following mathematical relations:

Normal distribution:

$$f(r) = \frac{1}{\sqrt{2\pi} \sigma} e^{-\frac{(r-r_0)^2}{2\sigma^2}}$$ (2.13)

Lognormal distribution:

$$\frac{1}{\sqrt{2\pi}} \frac{1}{r \ln(\sigma)} e^{-\frac{\ln^2(r/r_0)}{2\ln^2(\sigma)}}$$ (2.14)

where $r_0$ is the mode radius of the distribution and $\sigma$ is the width of the distribution. Figure 2.10 exhibits the shape of both type of PSD with a 60 nm mode radius and a width of 24 nm and 1.4 for the normal and lognormal distribution, respectively. Also shown is the contribution of the different portions of the PSD to the total effective scattering cross-section. The tail of larger particle sizes significantly contributes to the overall scattering, even though it represents only a small part of the entire particle population. On the other hand, particles with $r < r_0$ contribute less than 5% (for these particular cases) but constitute a large part of the entire ensemble.

Emission

For the sake of completeness, it is important to note that other than absorption and scattering, molecules and atoms in an excited state formed by processes resulting from solar illumination
can emit radiation. This phenomenon is called airglow. A number of species have emission features, the most important being O/O$^+$, N/N$^+$, O$_2$, N$_2$/N$_2^+$, Na, NO, OH and NO$_2$. Most terrestrial airglow emissions originate from electronic and molecular transitions, although rotational-vibrational transitions for constituents in their electronic ground state can occur as well. Such is the case for the OH molecule.
The summer mesopause region

The previous chapter introduced some basics concerning the source of the radiation in the atmosphere as well as the interaction of this radiation with atmospheric constituents. This chapter shall explore in more detail the composition and structure of the atmosphere, with an emphasis on the mesopause region and its unusual temperature structure at solstice. Finally, the formation of NLC near the summer mesopause and their known interaction with other atmospheric phenomena will be outlined.

3.1 Structure and composition of the atmosphere

The atmosphere is a gaseous layer surrounding the earth, composed mainly of N$_2$ (78%), O$_2$ (21%) and Ar (1%). At a given altitude $z$, the atmospheric pressure is related to the density $\rho$ according to the hydrostatic equation

$$\frac{dP}{dz} = -g \cdot \rho$$

(3.1)

where $g$, the gravitational acceleration, and $\rho$ are both function of the altitude. The atmosphere can generally be treated as an ideal gas, meaning that it follows the equation of state

$$P = \frac{\rho R_g T}{M}$$

(3.2)
where $R_g$ is the ideal gas constant, $T$ is the temperature (in Kelvin) and $M$ is the molar mass. Combining eq. 3.1 and eq. 3.2, the relationship between pressure and altitude is derived and given by

$$P(z) = P_o e^{-z/H}$$  \hspace{1cm} (3.3)

where $P_o$ is the pressure at the surface and $H$ is the so-called scale height. It is common practice to assume a scale height of approximately 7.5 km near the earth’s surface, but it should be kept in mind that the value of $H$ is not constant and depends on the temperature. According to eq. 3.3, the atmospheric pressure drops exponentially with height, which means that approximately 90% of the atmospheric mass is found within the lowermost 16 km of the atmosphere.

These figures can be misleading however. Despite the small amount of gases other than $N_2$, $O_2$ or Ar and the relative scarcity of atmospheric mass above 16 km, trace gases found in higher regions of the atmosphere can play exceedingly important roles regarding the energetics and chemistry of the atmosphere. It is well known for instance that $O_3$, representing less than $10^{-5}\%$ of the atmospheric composition, plays a critical role on our planet, shielding life on earth from harmful short-wave UV radiation through absorption in the upper layers of the atmosphere. Such interactions between radiation and composition are at the core of the vertical temperature structure of the atmosphere.

A sketch of a typical temperature profile is presented in figure 3.1. The atmosphere is commonly divided into four distinct layers: the troposphere, the stratosphere, the mesosphere and the thermosphere. These layers are defined by the different values of the vertical temperature gradients (also called lapse rates) in these regions, as can be seen from figure 3.1. The different layers are separated by transition regions where the temperature gradient changes rapidly: the tropopause, stratopause and mesopause, respectively.

There are basically three maxima in the temperature vertical profile, corresponding to hotspots of solar radiation absorption. The maximum in temperature in the troposphere is caused by the absorption of a large portion of the solar radiation by the surface, warming the overlying atmosphere. The temperature decrease is due to abundant convection taking place near the surface which provides for a well-mixed atmosphere (hence the name rooted in the Greek tropos for mixing, turning). When the vertical temperature gradient decreases below 2 K/km in magnitude, the troposphere gives way to the tropopause which acts as a cold trap for water vapor and significantly reduces the water content in the upper layers of the atmosphere through freeze drying.

The stratosphere is the layer found between about 15 and 50 km. It contains approximately 90% of the $O_3$ which, through absorption of short wavelength radiation, heats the atmosphere and is responsible for the temperature increase with altitude in this region. This temperature structure inhibits vertical mixing; consequently, the residence time of some constituents in this region are of the order of years. Many important chemical reactions take place in the stratosphere, mostly impacting ozone through catalytic cycles involving nitrogen, chlorine, bromine and hydrogen species. The excited species O($^1D$) also plays
3.1 Structure and Composition of the Atmosphere

Figure 3.1: U.S. standard atmosphere temperature profile. Taken from Brasseur and Solomon (2005).

a very important role in the stratosphere since it contributes to the destruction of some of the tropospheric source gases involved in the catalytic cycles affecting ozone such as CFCs. Another important chemical process which starts taking place around 30 km is the production of water vapor through oxidation of methane. This plays a large role in the formation of noctilucent clouds, since roughly half of the water content in the upper layer of the atmosphere is a result of the oxidation of methane (Thomas et al., 1989).

The mesosphere, situated above the stratosphere, is characterized by a decrease of temperature with altitude, a consequence of its relatively low O$_3$ concentration. At about 70 km, the methane oxidation reaction has essentially converted all the available CH$_4$ into H$_2$O, H and negligible amounts of odd-hydrogen radicals. The chemistry of the mesosphere can, to a large extent, be described by odd oxygen and odd hydrogen chemistry (Sonnemann and Grygalashvily, 2005). The most important reactions in the mesosphere involve the production of odd oxygen species through photolysis of molecular oxygen and ozone production as a result of a three-body reaction including atomic oxygen. The vast majority of the energy absorbed in the mesosphere is accounted for by molecular oxygen and ozone, while energy losses are due to radiative cooling by CO$_2$. The mesopause region, found between 88 and 100 km depending on the season and latitude, is the coldest place on earth with temperatures which can be as low as 130 K (Lübken, 1999). It is interesting to note that the lowest temperatures at the mesopause are reached during summertime, not wintertime.
A detailed explanation will be given in section 3.2.

The next layer above the mesopause is the thermosphere, a layer which possesses a positive vertical temperature gradient due to absorption of solar short-wavelength radiation by molecular oxygen. This highly energetic radiation breaks apart even strong bonds, which implies a decrease in the proportion of the dominant \( \text{O}_2 \) and \( \text{N}_2 \) molecules and consequently an increasing dominance of atoms. In this region, a transition from a well-mixed atmosphere to an environment where the vertical distribution of constituents is increasingly dependent on diffusive processes can be observed. The temperature in the thermosphere can reach values as high as 1000 K and is strongly dependent on solar activity. It should finally be noted that ion reactions are important in this part of the atmosphere and can significantly influence the neutral chemical composition.

### 3.2 The meridional circulation

It has already been mentioned that the polar summer mesopause is actually colder than the polar winter mesopause. Intuitively, with the summer pole being sunlit while parts of the winter pole are in the dark, it would be expected that the summer mesopause, through absorption of solar radiation, is warmer than its winter hemispheric counterpart. Figure 3.2a shows the temperature structure of the stratosphere and mesosphere assuming that radiative equilibrium is reached. It can be clearly observed that under these assumptions, the summer pole mesopause region actually features higher temperatures, on the order of 200 K, compared with the winter pole temperatures which can be as low as 130 K. In reality however, the opposite is the case (see figure 3.2b), which is the consequence of atmospheric dynamics driving temperature far from radiative equilibrium.

A simple understanding of the dynamical processes taking place in this region of the atmosphere can be gained by studying more closely the distribution of temperatures presented in figure 3.2b. It can be seen that the temperature is mainly latitude dependent, with the highest values at the summer stratopause around 50 km, due to strong ozone absorption, and decreasing gradually towards the winter pole. This meridional temperature gradient will give rise to a zonal wind gradient according to the thermal wind equation,

\[
f \frac{\partial u}{\partial z} = -\frac{R_g}{H R_E} \frac{\partial T}{\partial \phi}
\]  

\( (3.4) \)

where \( f \) is the Coriolis parameter, \( u \) the zonal wind velocity, \( z \) the log-pressure vertical coordinate, \( R_E \) the earth’s radius and \( \phi \) the latitude. The Coriolis parameter is defined as

\[
f = 2\omega \sin(\phi)
\]  

\( (3.5) \)

with \( \omega \) being the angular velocity of the earth. The resulting zonal flow, which is basically a consequence of the geostrophic balance between the Coriolis force and the pressure gradient,
3.2 THE MERIDIONAL CIRCULATION

Figure 3.2: Panel A: Meridional distribution of the modeled radiative equilibrium temperature in the stratosphere and mesosphere as calculated by Fels (1985). The dashed line indicates the lowest altitude shown in panel B. Panel B: MLS meridional temperature distribution for January.

is illustrated in figure 3.3. The zonal wind is westward in the summer hemisphere and eastward in the winter hemisphere, as calculated from eq. 3.4 and from the temperature distribution at solstice.

However, this geostrophic balance is disturbed by momentum deposition from internal gravity waves. Gravity waves are oscillations arising in a stably stratified atmosphere when an air parcel is displaced vertically. This type of waves is generated in the troposphere mainly by weather disturbances or by orography, and can propagate vertically with eastward or westward phase speed. The background zonal wind however has a filtering effect on the vertical propagation of these waves, as it absorbs gravity waves with a phase speed equal to its own background wind speed. Only waves with a phase speed opposite to (or larger than) the zonal wind can propagate higher in the atmosphere. Since in the summer stratosphere and mesosphere, the zonal wind is westward, mostly gravity waves with eastward phase speed will be able to penetrate higher in the atmosphere. As the atmospheric density decreases with height, the amplitude of the waves will grow accordingly up to the point where they become unstable and break, which effectively deposits energy and momentum in the atmosphere. Since the phase speed of the waves breaking is mostly eastward, the momentum deposited will act against the prevailing zonal wind and cause what is commonly known as wave drag. This situation however creates a geostrophic imbalance which is remediated by a meridional component of the wind, the direction of which is from the summer pole to the equator. At the winter pole, the reverse situation takes place: mostly westward gravity waves will break in the mesosphere, decelerating the zonal wind and creating a geostrophic imbalance resolved by a meridional wind component directed from the equator toward the winter pole. Therefore, there is a summer to winter pole circulation. Figure 3.4 shows the meridional distribution of the gravity wave momentum forcing in the
Figure 3.3: The distribution of zonal wind during solstice (July) from CIRA (COSPAR). The units of the zonal wind are m/s. Eastward winds are positive.

Figure 3.4: Meridional distribution of the gravity wave momentum forcing in the atmosphere (positive is eastward) as well as the pole-to-pole wave-driven circulation in the mesosphere (solid lines). Figure taken from Brasseur and Solomon (2005).

Combining this pole-to-pole circulation with mass continuity arguments, it ensues that upwelling takes place in the summer hemisphere and sinking in the winter hemisphere. As the air from lower altitudes at the summer pole is advected in the mesosphere, it expands
adiabatically due to the decrease in pressure with altitude, and effectively cools down. At the winter pole, an air parcel advected towards regions of higher pressure will experience a temperature increase. These processes are extremely effective in terms of heating and cooling, which explains the cold summer mesopause and the comparatively warmer winter mesopause, far away from radiative equilibrium. Figure 3.5a displays the daily mean MLS temperatures in the latitude range 70-80° between 40-90 km for year 2008, and the decrease in temperature at the mesopause during the summer months is readily apparent. Since the temperatures are very low, even in the winter mesopause, the response of infrared cooling to these large temperature perturbations is weak. This is the consequence of the Stefan-Boltzmann law, which states that the radiative cooling rate is proportional to $T^4$. Radiative cooling therefore not damp the large temperature differences effectively, hence the cold summer mesopause. The mean meridional circulation does not only have repercussions on the temperature, but also on the composition of the upper mesosphere. Constituents which are more abundant in the lower atmosphere are advected during summertime, leading to an increase in concentration. This is particularly the case for water vapor, a phenomenon which is presented in figure 3.5b displaying the MLS water vapor content for year 2008, similarly to figure 3.5a.

### 3.3 Summer mesopause

Because of the action of the meridional circulation, the summer mesopause is host to extremely low temperatures ($\sim$130 K) and an enhanced water vapor content ($\sim$6 ppm) which can result in saturation ratios in the range of 10-100 (Lübken, 1999). These conditions are favorable to the formation of ice particles and hence, noctilucent clouds. It has been shown by Hervig et al. (2001) that the main constituent of NLC particles is H$_2$O.

According to current theory, it is unlikely that NLC particles can form by homogeneous nucleation, at least via a direct transition from gas phase to ice. Zasetsky et al. (2009) recently suggested that, provided the process takes place in several distinct steps with reduced energy barriers, homogeneous nucleation could be thermodynamically possible. However, the presence of pre-existing cores could significantly lower the nucleation barrier for NLC particles, which makes heterogeneous nucleation more probable (Keesee, 1989). Several nucleation cores have been proposed, such as large proton hydrate ion clusters (Witt, 1969), sulfuric acid aerosol particles (Mills et al., 2005), sodium bicarbonate (Plane, 2000) and meteoric smoke particles (Rosinski and Snow, 1961; Hunten et al., 1980).

Meteoric smoke, the result of polymerization of metal compounds and silicon oxides (Kalashnikova et al., 2000), has been an especially popular candidate as a nucleation core since there is a constant input of material from meteoroids in the region of NLC formation. However, investigations by Megner et al. (2008a) using a two-dimensional model showed that the meridional circulation advects a large portion of the meteoric smoke particles away from the summer pole. This implies that the number densities of meteoric smoke particles
found near the summer mesopause are on the order of 100 particles cm\(^{-3}\), which is not consistent with what is expected for NLC formation (Megner et al., 2008b). A possible alternative to neutral particles is to consider charged meteoric smoke, which has been shown to be a reasonable candidate (Megner and Gumbel, 2009).

It is now known that NLC are actually the visible part of a thicker layer also containing smaller particles (Rapp et al., 2003; Rapp and Lübken, 2004) which can produce very strong radar echoes called Polar Mesosphere Summer Echoes (PMSE). These particles usually grow up to typical sizes of several tens of nanometers (Gumbel and Witt, 2001; Baumgarten et al., 2008; Robert et al., 2009) by direct deposition of water vapor on their surface. At such sizes, they can scatter light efficiently and become visible to optical instruments as well as ground observers under certain conditions. An ensemble of such particles forms a layer of roughly 1 km in vertical extent at altitudes of about 83 km (Fiedler et al., 2003) and has an ice particle density on the order of \(10^2\) cm\(^{-3}\) at the cloud peak brightness (Baumgarten et al., 2007). NLC particles are transported by winds and settle under the action of gravity, and eventually sublime when reaching subsaturated regions. The shape of the particles is customarily assumed to be spherical, but recent studies suggest that spheres cannot fully explain the NLC optical properties (Rapp et al., 2007; Baumgarten and Thomas, 2006) and that spheroid-shaped particles with axis ratios between 1/10 and 10 (Baumgarten et al., 2007) are more appropriate to account for the experimental data.

A typical NLC season starts about 3 to 4 weeks before the summer solstice and lasts for approximately 3 months (Olivero and Thomas, 1986). The occurrence rate increases until it reaches its maximum between day 15-20 relative to solstice, and decreases in a similar fashion. NLC appear preferably at higher latitudes where the temperature is lower and the water vapor mixing ratio higher. The occurrence rate at the season’s maximum can reach 90-100% at 80°N, 60-70% at 70°N and 10-20% at 60°N (Bailey et al., 2005). The higher temperatures at the southern summer mesopause, reported to be 4-7 K larger (Hervig and
Siskind, 2006; Wrotny and Russell, 2006; Lübken and Berger, 2007), make it a less hospitable environment for NLC, which explains the reduced maximum occurrence rates observed. More specifically, the maximum value of the southern hemisphere occurrence rate is about 80-90% at 80°S, 30-50% at 70°S and 5-10% at 60°S (Bailey et al., 2007). Other differences between both hemispheres include 35-40% brighter NLC and a 1-3 km lower cloud peak altitude in the north compared to the south (Petelina et al., 2006; Chu and Gardner, 2003).

Several processes affecting NLC on different timescales have been identified. Gravity waves propagating vertically up to the mesosphere can interact directly with NLC (Witt, 1961; Gerrard et al., 2004; Chandran et al., 2009a). It was first suggested by Hines (1968) that the structures observed in NLC could be the manifestation of interaction with internal gravity waves. Observation of gravity waves in NLC by Chandran et al. (2009a) showed the presence of waves with horizontal wavelengths in the range 15-320 km. New studies also show that gravity waves are likely to be the source of the zonal variation in NLC occurrence frequency, due to the substantial momentum and energy deposition by gravity waves in this region.

Planetary waves are also known to affect NLC occurrence and brightness through a modulation of temperature. The most common planetary waves signature found in an analysis of NLC properties is the 5-day wave (Kirkwood and Stebel, 2003; Merkel et al., 2003; von Savigny et al., 2007a), but the 2-day wave is also present (Merkel et al., 2008), especially in the SH where it is known to be active in the second half (January, February) of the NLC season (Morris et al., 2009).

Diurnal variation in NLC properties has been shown to be linked to atmospheric tides (von Zahn et al., 1998). An investigation of the seven years lidar NLC data set shows a recurrent pattern in the variation of the NLC properties over the diurnal cycle, which also suggests that this effect is the result of atmospheric tides (Fiedler et al., 2005).

Karlsson et al. (2007) established that the dynamical coupling between the summer mesosphere and the winter stratosphere (Becker and Fritts, 2006), the so-called interhemispheric coupling, significantly affects interannual variability of NLC properties. A recent study by Karlsson et al. (2009) shows that the process is also efficient at modulating NLC properties within the season. The time lag between changes in the winter stratosphere and the connected response in NLC properties in the summer mesosphere is found to be between 2 and 8 days.

It was also argued that the lunar cycle could affect the NLC occurrence rate (Dalín et al., 2006) due to the change in earth-moon distance. Non-periodic events can also have an impact on NLC properties, like solar proton events (SPE) (von Savigny et al., 2007b), space shuttle launches (Stevens and Englert, 2005) and possibly volcano eruptions (Thomas and Olivero, 2001).

The 11-year solar cycle is known to appreciably modulate NLC properties (Gadsden, 1985; Thomas et al., 1991). DeLand et al. (2003) assessed the impact of the 11-year solar cycle...
cycle on NLC activity using more than 25 years of NLC measurements, showing significant anti-correlation between the NLC albedo and the Lyman-α irradiance in both hemispheres, with a stronger anti-correlation in the northern hemisphere. Figure 3.6 shows the clear anti-correlation between NLC albedo and the Lyman-α variation for the period 1979-2004 for both hemispheres. Hervig and Siskind (2006) confirmed these findings and showed that the variation in NLC properties was most probably a consequence of temperature and water vapor changes in the upper mesosphere. Moreover, once the effect of the solar activity is removed from the long Solar Backscattering UltraViolet (SBUV) instruments’ time series, a positive secular trend of up to 20% was observed over the last 27 years in both NLC occurrence and albedo (Shettle et al., 2009; DeLand et al., 2007). Another indication of a change in NLC occurrence patterns is the recent ground observation of night-shining clouds at latitudes as low as 42° (Taylor et al., 2002), although it would be premature to draw conclusions based on the very few sightings available. It has been argued that this long-term change in NLC properties could be caused by an enhanced radiative cooling of the upper atmosphere due to a rise in greenhouse gas concentration as well as an increase in mesospheric water vapor concentrations (Olivero and Thomas, 2001; Grygalashvyly and Sonnemann, 2006). Most measurements, however, do not support the hypothesis of a significant long-term temperature decrease near the mesopause (Lübken, 2000; Beig et al., 2003), and between 1992 and 2002, a decrease in H₂O content at NLC height was observed (von Zahn et al., 2004).

Finally, NLC are not merely bystanders to these abundant atmospheric processes taking place in the vicinity of the polar summer mesopause, but can also play an active role in the chemistry of the upper mesosphere. As NLC represent a large reservoir of water mostly
unavailable in vapor form at NLC heights, the particle sedimentation acts as a mechanism for the vertical redistribution of water. The ice particles settling through the atmosphere reach the threshold of saturation and sublimate, producing a layer of enhanced water vapor below the cloud, while the region above the NLC is depleted in water vapor. This depletion of H₂O above the cloud has been shown to enhance the ozone content by about 20-30%, a phenomenon predicted by modeling and confirmed by satellite observations (Siskind et al., 2007). The redistribution of water in the polar upper mesosphere due to the presence of NLC can also lead to a decrease in atomic hydrogen in the lowermost thermosphere (Siskind et al., 2008). Furthermore, NLC are known to interact with metallic species of meteoric origin. NLC are great scavengers of iron (Plane et al., 2004), sodium (She et al., 2006) and potassium (Lübken and Höffner, 2004), completely removing these constituents from the atmosphere due to the efficient uptake of these species on ice particle surfaces.
Instruments and data sets

The results of this work are based on a number of different instruments and available data sets. First and foremost, the retrieval of NLC properties, which will be described in chapter 5, is based on measurements made by SCIAMACHY. This instrument will be described thoroughly in the first part of this chapter, since it is necessary to understand the characteristics and limitations associated with SCIAMACHY and to take them into account in the analysis of its measurements. Subsequent sections will describe the important features of the various data sets exploited throughout this work, as well as the uncertainties associated with them.

4.1 SCIAMACHY

In this work, the retrieval of NLC properties is derived from spectrometric data gathered by the SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY, or more simply SCIAMACHY. This instrument is the outcome of a fruitful collaboration between Germany, the Netherlands and Belgium after more than a decade in the making. SCIAMACHY (Goede et al., 1991; Burrows et al., 1995; Bovensmann et al., 1999; Gottwald et al., 2006) is part of the ambitious Environmental Satellite (ENVISAT) mission, developed by the European Space Agency (ESA) with the goal of gaining insights into local and global environmental processes taking place on land, in the ocean, in the cryosphere as well as in the atmosphere. The launch of this large satellite (26 m × 10 m × 5 m) - the largest ever built by ESA - took place on March 1, 2002 in Kourou, French Guyana. A depiction of Envisat in orbit is presented in figure 4.1a. The 2050 kg payload comprises 10 instruments
in total, 7 of which were developed by ESA itself and 3 more being so-called *Announcement of Opportunity* instruments which are provided by national agencies and developed under their responsibilities. SCIAMACHY is one of these instruments, besides AATSR and DORIS. The satellite orbits the earth at a height of 799.8 km in a polar sun-synchronous orbit, with an equator descending crossing-time (DNX) of 10:00 local time (LT). As it journeys to higher latitudes ($\gtrsim 70^\circ$), the sub-satellite point quickly covers different local time zones. Figure 4.1b shows the Envisat overflight local time for different latitudes. It takes 100.6 minutes to circle the earth, which means that every day, the satellite revolves around the planet about 14 times.

The Envisat mission was originally planned for 5 years, but due to its excellent performance and the high data demand from both the science and operational user communities, a mission extension until at least 2013 has been unanimously approved by the ESA Earth Observation Programme Board. The limiting factor for the mission now lies in the amount of available fuel (hydrazine) onboard the spacecraft, which is necessary for orbit control manoeuvres. The implementation of new orbital parameters minimizing the fuel consumption is supposed to take place in October 2010.

The motivation behind the SCIAMACHY instrument was to determine concentrations of a large number of trace gas species over the full vertical extent of the atmosphere. Achieving this goal requires that the instrument continuously observes radiation with wavelengths from the UV to the IR region. SCIAMACHY is a passive remote sensing instrument using the sun and the moon as source of electromagnetic radiation. Its radiation measurements actually cover the wavelength range 214 nm to 2386 nm, albeit non-continuously for $\lambda > 1750$ nm. The incoming radiation is split in 8 channels, each having its own spectral resolution which varies between 0.24 and 1.48 nm. Details on SCIAMACHY spectral coverage and resolution can be found in table 4.1.

What makes SCIAMACHY a pioneering instrument are its 3 different modes of observation: nadir, limb and occultation. Figure 4.2a shows SCIAMACHY’s geometry of observation with
4.1 SCIAMACHY

<table>
<thead>
<tr>
<th>Channel</th>
<th>Wavelength [nm]</th>
<th>Resolution [nm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>212-314</td>
<td>0.21</td>
</tr>
<tr>
<td>2</td>
<td>309-404</td>
<td>0.22</td>
</tr>
<tr>
<td>3</td>
<td>392-605</td>
<td>0.47</td>
</tr>
<tr>
<td>4</td>
<td>598-790</td>
<td>0.42</td>
</tr>
<tr>
<td>5</td>
<td>776-1056</td>
<td>0.55</td>
</tr>
<tr>
<td>6</td>
<td>991-1750</td>
<td>1.56</td>
</tr>
<tr>
<td>7</td>
<td>1940-2040</td>
<td>0.21</td>
</tr>
<tr>
<td>8</td>
<td>2261-2380</td>
<td>0.24</td>
</tr>
</tbody>
</table>

Table 4.1: SCIAMACHY spectral coverage and resolution for channels 1-8.

Figure 4.2: Panel A: SCIAMACHY observation modes and their physical characteristics. PanelB: Nominal SCIAMACHY observation sequence during one orbit. Adapted from Gottwald et al. (2006).

regard to the earth while performing the different types of measurements.

When performing nadir measurements, radiation enters the nadir port and is redirected inside the instrument by a scanning mirror, the elevation scan mechanism (ESM). The ESM scans the sub-satellite air masses $\pm 32^\circ$ across-track. Although the scanner can in principle rotate $360^\circ$ around its axis, the baffles limit the scanning at large angles. The entrance slit of the spectrometer defines an Instantaneous Field of View (IFoV) of $1.8^\circ \times 0.045^\circ$ which corresponds to a ground pixel size of 25 km $\times$ 0.6 km at the sub-satellite point. The spatial
resolution obtained for nadir measurements depends on the scanning velocity and the integration time of the measurement and can range between 26 km × 30 km (along-track × across-track) up to 32 km × 930 km. A typical spatial coverage of SCIAMACHY nadir measurements during one orbit is presented in the left panel of figure 4.3. At present, nadir products retrieved at the Institute of Environmental Physics in Bremen (IUP) include O₃, NO₂, BrO, HCHO, SO₂, OClO and H₂O, CH₄, CO, CO₂, N₂O as well as tropospheric clouds and aerosols properties.

Limb measurements provide information on the vertical distribution of atmospheric constituents and properties. In this geometry, the instrument’s line-of-sight (LOS) goes through the earth’s limb, and photons scattered along this LOS are measured some 3000 km away from the LOS tangent point. The incoming radiation goes through the limb port, onto the azimuth scanning mechanism (ASM) mirror which redirects it to the ESM where it is finally led inside the spectrometer. The horizontal scanning is done through the ASM while changes in the ESM position are responsible for the vertical scanning. The instrument’s baffles restrict the scanning of the limb to ±44° across-track, but vertically it can scan from the ground up to 380 km above the earth’s surface. The limb IFoV is 105 km × 2.5 km at the tangent point. The spatial resolution ranges from 1060 km × 400 km × 3.6 km (across-track × along-track × vertical) to 230 km × 400 km × 2.6 km, determined by the integration time. The instrument scans the atmosphere at 31 tangent heights (TH) in steps of 3.3 km, usually from the ground up to 90 km in nominal mode plus a measurement at about 270 km. Currently retrieved products from limb measurements comprise O₃, BrO, NO₂, OClO, Mg species and clouds and aerosols properties. The spatial coverage of SCIAMACHY limb measurements for orbit 28009 (July 9, 2007) is shown in the right panel of figure 4.3. Since NLC occur polewards if 50°, it is important to note that a typical daily limb coverage at high latitudes during the summer season comprises between 20-30 measurements per 5° latitude bin for both hemispheres. This coverage includes the ascending and the descending portion of the orbit.
Occultation measurements take place when the sun or the moon are present in the field of view (FoV) of the instruments during orbital sunrise or moonrise. Measurements in this observation mode have the advantage of being almost self-calibrating so that retrievals are immune to long-term instrumental degradation. In addition, they have a large signal-to-noise (SN) ratio because of the bright sources that the sun and the moon represent. However, they suffer from poor spatial coverage since there can be only one sunrise/moonrise per orbit. Solar occultation can only take place at latitudes polewards of 65°N. Occultation of the moon covers the southern hemisphere, usually for latitude southwards of 60°S. The vertical resolution for solar occultation measurements is about 30 km × 400 km × 2.6 km and is very similar for lunar occultation, except for the vertical resolution which is about 3 km.

Aside from these 3 measurement modes, SCIAMACHY can also measure solar irradiance via the subsolar port, where it is measured via a diffuser. Two aluminum diffusers are available: one on the backside of the ESM mirror and another on the backside of the ASM mirror. The optimized ASM diffuser was added after calibration on the ground had shown that the ESM diffuser exhibited spectral features which would have endangered successful retrieval of some trace gas species.

All of these modes of observation are done in a given sequence for every orbit, as shown in figure 4.2b. A typical SCIAMACHY orbit starts with 4 consecutive limb measurements during twilight, followed by solar occultation and then a succession of limb and nadir measurements. One interesting aspect of this procedure is that the limb and nadir measurements are optimized to measure overlapping air masses, and combining both measurements can lead to a determination of tropospheric vertical column of trace gases of interest (Sierk et al., 2006). If moon occultation is possible, such measurements are performed every second orbit. The rest of the sequence is usually used for calibration purposes and solar irradiance measurements through the sub-solar port.

**Optical Design**

The radiation collected by SCIAMACHY in the various geometries of observation is invariably directed by the ESM mirror in the optical assembly, organized in two levels, which generates the spectral information as output (see figure 4.4). The incoming radiation is focused on the spectrometer entrance slit by a telescope, and continues its path to a pre-dispersive prism. The main beam leaving this prism is then separated into four beams which contain light for channels 1, 2, 3-6 and 7-8 respectively. All of these rays are then further isolated in the appropriate channels, where they are finally dispersed by a grating onto a diode array detector containing 1024 pixels. This partition of radiation within the instrument has the advantage of minimizing spectral stray light in the channels covering low light intensity in the UV, near infrared (NIR) and short-wave infrared (SWIR) part of the spectrum. Entrance optics, pre-dispersive prism, the calibration unit and channels 1 and 2 are contained in level
1, facing in the flight direction, while channels 3 to 8 are located in level 2.

It should be kept in mind that since SCIAMACHY is a grating spectrometer without a polarization scrambler, its measurements are sensitive to the polarization of the incoming light. Measurement of the polarization is done for 6 broadband polarization channels (PMD), roughly corresponding to channels 2-6 and channel 8.

In order to minimize detector noise and dark current, the diode arrays are cooled to temperatures ranging from 150 to 235 K. The entire optical bench is also kept at 253 K to minimize the IR emission which could potentially interfere with measurements from channel 6 to 8. Level 1 contains two light sources for calibration purposes. The NePtCr hollow cathode discharge lamp emits light with very narrow lines so as to permit spectral calibration of the instrument. The 5 W Tungsten white lamp, on the other hand, is used to monitor pixel to pixel gain differences as well as long-term degradation of the optical components. This is crucial for the in-flight calibration and monitoring of the instrument (Noël et al., 2003). Information gathered either during the monitoring and calibration procedure in space or on the ground before the launch makes it possible to correct the radiometric quantities, adjusting them for undesirable effects such as the memory effect, non-linearity of the detector and stray light correction.

One aspect of SCIAMACHY’s monitoring is especially relevant for this project as it is connected with channel 1 which is used for the retrieval of NLC properties (von Savigny et al., 2007c). Since the beginning of the mission, degradation of channel 1 is taking place, as shown in figure 4.5. One notices that the degradation affects shorter wavelengths more strongly at any given time after 2004, incorporating a spurious change in the variation of measured radiance with wavelength. As of October 2009, the transmission is as low as 20% for $\lambda = 240$ nm. Moreover, the transmission does not decrease linearly with time. The
impact of the degradation on the determination of the NLC particle size will be discussed more exhaustively in section 5.5.

The monitoring factors (M-factors) database (Bramstedt, 2008) is a useful tool developed by Klaus Bramstedt and Stefan Noël at IUP Bremen to adjust for the degradation of SCIAMACHY’s optical components. One of the great advantages is that these M-factors can be applied during the data processing from L1b to L1c level by the data users, which means that the database is decoupled from the operational 0-1b processing and the corrections can be applied “online”. The M-factor corrections are also more reliable (Stefan Noël, pers. comm.) than the degradation dataset available from the SCIAMACHY Operations Support Team (SOST) (http://www.iup.uni-bremen.de/sciamachy/LTM/LTM.html). In this work, the M-factors database version 6.01 available online (http://www.iup.uni-bremen.de/sciamachy/mfactors/) is used.

4.2 Data sets

SBUV and SBUV/2 instruments

The Solar Backscatter Ultraviolet instrument (SBUV) is a spectrally scanning sounding radiometer designed primarily to measure atmospheric ozone on a global scale. The first iteration of the instrument was introduced in 1978 on-board the Nimbus-7 (Cebula et al., 1988). An improved version (SBUV/2) was later part of the NOAA-9, NOAA-11, NOAA-14, NOAA-16, NOAA-17, NOAA-18 and NOAA-19 platforms so that, altogether, more than 30 years of operational data are available.
The instrument is designed to measure the earth radiance as well as the solar irradiance at 12 discrete wavelengths between 160 to 406 nm. The presence of a programmable grating allows the modification of the wavelength measured by the different channels of the instrument, but standard measurements are done at wavelengths between 252 and 340 nm. Table 4.2 details the central wavelengths nominally measured by the SBUV/2-type instruments.

Although primarily employed to retrieve ozone profiles, Thomas et al. (1991) used the information available in the first 5 SBUV channels (252.0-292.3 nm) to detect NLC events and determine their albedo, defined as the ratio of the measured NLC radiance and the solar radiance at a given wavelength. As explained by DeLand et al. (2003), NLC can be identified in the SBUV data set due to their enhanced scattering of UV radiation, which has a different wavelength dependance from a decrease in ozone.

The procedure to detect NLC in the SBUV data set requires first the calculation of the daily background albedo for all wavelengths. This is done by fitting a 4th order polynomial through the absolute albedo as a function of the solar zenith angle (SZA). The residuals of the albedos are found by simply subtracting the daily background from the absolute albedo. A typical background fit for the 252 nm albedos from NOAA-9 is presented in figure 4.6a.

### Table 4.2: SBUV/2 nominal channel central wavelengths and bandwidths. Adapted from Robel (2009).

<table>
<thead>
<tr>
<th>Channel</th>
<th>Central Wavelength [nm]</th>
<th>Bandwidth [nm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>252.00 ± 0.05</td>
<td>1 ± 0.2</td>
</tr>
<tr>
<td>2</td>
<td>273.61 ± 0.05</td>
<td>1 ± 0.2</td>
</tr>
<tr>
<td>3</td>
<td>283.10 ± 0.05</td>
<td>1 ± 0.2</td>
</tr>
<tr>
<td>4</td>
<td>287.70 ± 0.05</td>
<td>1 ± 0.2</td>
</tr>
<tr>
<td>5</td>
<td>292.29 ± 0.05</td>
<td>1 ± 0.2</td>
</tr>
<tr>
<td>6</td>
<td>297.59 ± 0.05</td>
<td>1 ± 0.2</td>
</tr>
<tr>
<td>7</td>
<td>301.97 ± 0.05</td>
<td>1 ± 0.2</td>
</tr>
<tr>
<td>8</td>
<td>305.87 ± 0.05</td>
<td>1 ± 0.2</td>
</tr>
<tr>
<td>9</td>
<td>312.57 ± 0.05</td>
<td>1 ± 0.2</td>
</tr>
<tr>
<td>10</td>
<td>317.56 ± 0.05</td>
<td>1 ± 0.2</td>
</tr>
<tr>
<td>11</td>
<td>331.26 ± 0.05</td>
<td>1 ± 0.2</td>
</tr>
<tr>
<td>12</td>
<td>339.89 ± 0.05</td>
<td>1 ± 0.2</td>
</tr>
<tr>
<td>Cloud Cover</td>
<td>379.00 ± 1</td>
<td>3 ± 0.3</td>
</tr>
</tbody>
</table>
while panel B shows the same data but in terms of residual albedo. The residual albedo of a NLC event at a given wavelength $r_\lambda$ must fulfill four conditions:

1. $r_{252}, r_{273}, r_{283} > 0$
2. The slope of the linear fit of $r(\lambda)$ vs $\lambda$ must be negative.
3. $r_{252}$ must be larger than a noise term determined partly by the daily variability of the residual albedo.
4. $r_{252} > r_{273}$

This procedure is iterated five times, with each new iteration excluding detected NLC events from the data for the background fit calculation. The data set produced in this way allows for the calculation of the NLC albedo for a single event or an ensemble of events. Section 3 of the paper by DeLand et al. (2007) discusses the uncertainty of the 252 nm albedo for a single NLC detection and estimates it at 20-25%. However, since most of the uncertainty represents random variations, that number should decrease by a factor of $\sqrt{N}$ when averaging over $N$ detections. A reasonable estimate of the uncertainty of NLC albedo daily averages is around 3-4%. Another quantity which can be calculated is the NLC occurrence rate which is simply the ratio of the number of NLC events taking place in a given time interval and within a given spatial cover and the total number of measurements which took place in this time interval and within the spatial boundary defined.

An example of a daily map of NLC retrieved by a SBUV/2 type instrument is shown in figure 4.7. The pixel dimensions at the surface are 170 km $\times$ 170 km. More information about the NLC detection algorithm used to produce the data set employed in this work can be found in the article by DeLand et al. (2003).
MLS

The Microwave Limb Sounder (MLS) measures the thermal microwave limb emissions from atmospheric species in five spectral regions from 115 GHz to 2.5 THz (Waters et al., 2006). It is on-board the Aura (Latin for breeze) spacecraft, launched 15 July 2004 in a sun-synchronous orbit with a 13.45 LT ascending equator-crossing time. MLS scans the atmosphere vertically from the ground to \( \sim 90 \) km. Its vertical scan rate varies with altitude, being faster in the upper regions of the atmosphere and resulting in a coarser vertical resolution in the mesosphere. MLS Level 2 geophysical product which are going to be used in this work are temperature and water vapour content (version2.2). Geopotential heights (GPH) are also indirectly used to calculate the vertical distribution of these products on a geometric altitude grid instead of a pressure grid.

The retrieval of the temperature profile is based on measurements of the thermal emission of O\(_2\) near 118 and 234 GHz (Schwartz et al., 2008), while the H\(_2\)O is retrieved from the emissions at 183.31 GHz (Lambert et al., 2007). The resolution of a single temperature profile near the mesopause is about \( 15 \times 220 \times 6 \) km (vertical \( \times \) horizontal along-track \( \times \) horizontal cross-track) and has a 2.5 K precision in the mesosphere. Table 4.3 details the characteristics of the MLS temperature product.

Measurements of temperature and H\(_2\)O, among other species, are done using digital autocorrelator spectrometers with a resolution of 0.15 MHz to measure narrow spectral lines at atmospheric pressures below 1 hPa, important for measurements in the middle-atmosphere. The spatial resolution for MLS H\(_2\)O product in the mesosphere is about \( 13 \times 400 \times 7 \) km, with a 180% precision and a minimum measurable H\(_2\)O mixing ratio of 0.1 ppmv. The particulars of the MLS water vapour product are presented in table 4.4.
Table 4.3: Summary of MLS temperature product features. The precision and resolution relate to single profile retrievals while the observed scatter is derived from comparison of individual profiles from successive orbits. Adapted from Livesey et al. (2007).

<table>
<thead>
<tr>
<th>Pressure [hPa]</th>
<th>Precision [K]</th>
<th>Observed Scatter [K]</th>
<th>Resolution V × H [km]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.001</td>
<td>± 2.5</td>
<td>± 3.5</td>
<td>15 × 220</td>
</tr>
<tr>
<td>0.01</td>
<td>± 2.2</td>
<td>± 3</td>
<td>14 × 185</td>
</tr>
<tr>
<td>0.1</td>
<td>± 2</td>
<td>± 2.3</td>
<td>9.1 × 170</td>
</tr>
<tr>
<td>0.316</td>
<td>± 1</td>
<td>± 1.5</td>
<td>8.3 × 165</td>
</tr>
<tr>
<td>1</td>
<td>± 1</td>
<td>± 1.4</td>
<td>7.9 × 165</td>
</tr>
<tr>
<td>3.16</td>
<td>± 0.8</td>
<td>± 1</td>
<td>6.2 × 165</td>
</tr>
<tr>
<td>10</td>
<td>± 0.6</td>
<td>± 1</td>
<td>4.3 × 165</td>
</tr>
<tr>
<td>14.7</td>
<td>± 0.6</td>
<td>± 1</td>
<td>3.9 × 165</td>
</tr>
<tr>
<td>31.6</td>
<td>± 0.6</td>
<td>± 1</td>
<td>3.5 × 165</td>
</tr>
<tr>
<td>56.2</td>
<td>± 0.8</td>
<td>± 0.8</td>
<td>3.8 × 165</td>
</tr>
<tr>
<td>100</td>
<td>± 0.8</td>
<td>± 0.8</td>
<td>5.2 × 165</td>
</tr>
<tr>
<td>215</td>
<td>± 1</td>
<td>± 1</td>
<td>5.0 × 165</td>
</tr>
<tr>
<td>316</td>
<td>± 1</td>
<td>± 1</td>
<td>5.3 × 165</td>
</tr>
</tbody>
</table>

Lyman-α composite time series

Parts of this work deal with the interaction of solar activity with the upper atmosphere, especially the interaction on a 27-day timescale. As already indicated earlier in section 2.1, many different proxies of solar activity exist. In the present work, the Lyman-α flux will be used. The primary reason for this choice is the fact that the Lyman-α line is actually known to interact with the Earth’s atmosphere chiefly at altitudes between 70 and 110 km, where it dissociates oxygen and water vapour and ionizes nitric oxide to form the ionosphere’s D-layer. Moreover, it is superior to the ground-based solar 10.7 cm radio flux measurements because unlike the latter, it can still register the 27-day variation during quiet sun conditions (Barth et al., 1990).

Another reason for using the Lyman-α flux as a proxy is the availability of a time series which goes back to 1947. The data set was put together by Woods et al. (2000) by combining measurements from different satellite instruments and using predictions from proxy models to fill the gaps between the different space-borne missions. The uncertainty of the Lyman-α irradiance is estimated at 10% and is mainly due to the composite nature of the time series.
Table 4.4: Summary of MLS water vapour product characteristics for individual profiles. Adapted from Livesey et al. (2007).

<table>
<thead>
<tr>
<th>Pressure [hPa]</th>
<th>Precision [%]</th>
<th>Accuracy [ppmv]</th>
<th>Resolution V × H [km]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.002</td>
<td>180</td>
<td>34</td>
<td>13 × 320</td>
</tr>
<tr>
<td>0.004</td>
<td>82</td>
<td>16</td>
<td>13 × 360</td>
</tr>
<tr>
<td>0.010</td>
<td>34</td>
<td>11</td>
<td>12 × 390</td>
</tr>
<tr>
<td>0.022</td>
<td>18</td>
<td>9</td>
<td>12 × 420</td>
</tr>
<tr>
<td>0.046</td>
<td>10</td>
<td>8</td>
<td>16 × 430</td>
</tr>
<tr>
<td>0.10</td>
<td>6</td>
<td>8</td>
<td>14 × 440</td>
</tr>
<tr>
<td>0.22</td>
<td>5</td>
<td>7</td>
<td>6.7 × 420</td>
</tr>
<tr>
<td>0.46</td>
<td>4</td>
<td>6</td>
<td>5.5 × 410</td>
</tr>
<tr>
<td>1.00</td>
<td>4</td>
<td>4</td>
<td>4.6 × 410</td>
</tr>
<tr>
<td>2.15</td>
<td>4</td>
<td>5</td>
<td>4.0 × 380</td>
</tr>
<tr>
<td>4.64</td>
<td>4</td>
<td>7</td>
<td>3.6 × 320</td>
</tr>
<tr>
<td>10</td>
<td>4</td>
<td>9</td>
<td>3.3 × 280</td>
</tr>
<tr>
<td>22</td>
<td>4</td>
<td>7</td>
<td>3.2 × 220</td>
</tr>
<tr>
<td>46</td>
<td>6</td>
<td>4</td>
<td>3.5 × 180</td>
</tr>
<tr>
<td>68</td>
<td>8</td>
<td>6</td>
<td>3.4 × 180</td>
</tr>
<tr>
<td>83</td>
<td>10</td>
<td>7</td>
<td>3.3 × 180</td>
</tr>
<tr>
<td>100</td>
<td>15</td>
<td>8</td>
<td>3.5 × 200</td>
</tr>
<tr>
<td>121</td>
<td>20</td>
<td>12</td>
<td>3.3 × 185</td>
</tr>
<tr>
<td>147</td>
<td>20</td>
<td>15</td>
<td>3.5 × 200</td>
</tr>
<tr>
<td>178</td>
<td>25</td>
<td>20</td>
<td>3.3 × 185</td>
</tr>
</tbody>
</table>
The latest version of the data is available online from the Lasp Interactive Solar Irradiance Datacenter (LISIRD) at http://lasp.colorado.edu/lisird and is updated weekly using TIMED SEE and SORCE SOLSTICE solar irradiance measurements.
In this chapter, the NLC detection algorithm for SCIAMACHY will be presented. Using the information available in scattered radiation from NLC particles, it is possible to derive their size, provided we make some assumptions about the particle’s population distribution and shape. The algorithm used for the retrieval of SCIAMACHY’s NLC radiance and particle size will be presented along with an estimation of their uncertainties.

### 5.1 NLC observation geometry from SCIAMACHY

The retrieval of NLC properties from SCIAMACHY is done in limb observation geometry. The advantage of this measurement configuration lies in the long optical path through the cloud ensuring considerable scattering in the direction of the instrument. In addition, the measurements are done against a dark background leading to good signal to noise ratios. These characteristics are especially valuable if one compares them with measurements from a nadir viewing instrument, such as SBUV, for which the NLC detection must be performed against the bright background of the earth’s atmosphere and with an optical path through the cloud equivalent to the thickness of the NLC layer. As for any satellite platform, the large geophysical coverage and the fact that NLC can be detected at any time under sunlit conditions are also an asset compared to lidar measurements and ground-based photography. Besides the limb observation geometry, SCIAMACHY’s excellent spectral coverage and resolution are further advantageous attributes which play an important role in the determination of NLC particle size (as will be presented in section 5.5). The use of
UV radiation results in a high sensitivity of the NLC retrieval to a much larger part of the particle population, compared to visible radiation retrievals (Thomas, 1991).

However, NLC detection with limb observing instruments such as SCIAMACHY is not without limitations (Karlsson and Gumbel, 2005). The detection in this mode of observation makes it impossible to know whether the NLC detected is present at the tangent height, in the foreground or the background. This will contribute to higher uncertainty in their geographical distribution. The large volume observed by SCIAMACHY in nominal observation mode, 400 km (along-track) × 1000 km (across-track) × 3 km (vertical) inhibits the resolution of small-scale features of the clouds and leads to a large spatial smoothing of their properties. The sun-synchronous orbit of the satellite also implies that measurements at different latitudes are done at different local times, especially at polar latitudes (see figure 4.1). Bearing this in mind, it is difficult to uncouple the effect of each of these parameters on NLC properties.

A depiction of the observation of a NLC using SCIAMACHY is presented in figure 5.1. The NLC is modeled as an ice particle layer of thickness $h$ with bottom altitude $z_b$. The tangent height of the LOS of the instrument is $z_r$. The total radiance measured by the instrument is

$$L_T^{\lambda} = L^{NLC}_\lambda + L^{Rayleigh}_\lambda + \epsilon_\lambda$$  \hspace{1cm} (5.1)
Figure 5.2: Simulated sun-normalized limb radiances at selected tangent heights (left) and the relative contribution of single scattering, multiple scattering and surface reflection to the total measured radiance (right). Adapted from Kaiser and Burrows (2003)

where $L^\text{Ray}_\lambda$ is the contribution of the Rayleigh scattering due to the molecular background, $L^\text{NLC}_\lambda$ is the radiance attributable to scattering by NLC particles (with Mie-like properties in the UV) and $\varepsilon_\lambda$ is a term comprising instrumental noise, spatial and spectral stray light, possible atmospheric emission features or any other radiance source unaccounted for. The NLC scattered radiance $L^\text{NLC}_\lambda$ is computed by

$$L^\text{NLC}_\lambda = S_\lambda \int_{\text{FoV}} \int_{\text{LOS}} ds \int_0^{+\infty} dr \frac{d\sigma^\text{NLC}}{d\Omega} \cdot n^\text{NLC}(z) \cdot f(z, r) \cdot e \left( -\int_{-\infty}^{0} ds' \sigma^\text{NLC}(z(s')) \right)$$

(5.2)

where $S_\lambda$ is the solar irradiance at a given wavelength scattered by an ensemble of particles with a PSD $f(z, r)$ which varies with altitude and hence along the optical path $s$. Each particle has a differential scattering cross-section $d\sigma^\text{NLC}/d\Omega$ dependent on the scattering angle $\theta$, the particle size $r$ and the wavelength. The number density of scattering particles $n^\text{NLC}(z)$ varies along the LOS. The extinction of the scattered radiation along the LOS, either by scattering or absorption, is taken into account in the form of the exponential term. Only the scattered radiance within the FoV is measured by the instrument, so that the integral over the solid angle extends over the FoV. Since SCIAMACHY exploits UV radiation for NLC detection, it ensues that the strong absorption by O$_3$ at lower altitude removes any effect from the ground albedo on the retrieval, so that only solar irradiance is considered as a source of radiation. Moreover, it ensures that most of the measured scattered photons below 300 nm have only experienced single scattering (Kaiser and Burrows, 2003), as illustrated in Figure 5.2. This simplifies and speeds up the retrieval considerably since no complex radiative transfer model is required for the analysis of the measured limb radiation.
The expression for $L_{\text{Ray}}^{\lambda}$ has the same form as eq. 5.2

\[ L_{\text{Ray}}^{\lambda} = S_{\lambda} \int_{\text{FoV}} d\Omega \int_{\text{LOS}} ds \cdot \sigma_{\text{Ray}}(\lambda, \theta) \cdot n^{\text{Ray}}(z) \cdot e \left( -\int_{-\infty}^{s} ds' \sigma_{\text{n}}(z') \right) \] (5.3)

with $n^{\text{Ray}}(z)$ being the molecular number density at altitude $z$ and $d\sigma_{\text{Ray}}/d\Omega$ being the Rayleigh differential scattering cross-section.

Since extinction along the line of sight is negligible at NLC altitudes ($\tau < 10^{-2}$ according to Debrestian et al. (1997)), the exponential term in both expressions is set to 1. Moreover, we assume that the PSD and the particle number density do not vary with height so that $f(z, r) = f(r)$ and $n^{\text{NLC}}(z) = n^{\text{NLC}}$. Taking these assumptions into account, the expression for the NLC radiance becomes

\[ L^{\text{NLC}}_{\lambda} = S_{\lambda} \cdot n^{\text{NLC}} \int_{\text{FoV}} d\Omega \int_{\text{LOS}} ds \cdot \int_{0}^{+\infty} dr \cdot \sigma_{\text{NLC}}(\lambda, r, \theta) \cdot f(r) \] (5.4)

i.e. the NLC radiance is the product of the solar irradiance, the number of particles in the FoV and the effective differential scattering cross-section of the particle ensemble. The integral over the optical path through the layer can be calculated using simple geometrical considerations. The length of the LOS through the layer depends on the location of the tangent height of the LOS relative to the layer:

CASE A: $z_b > z_r$

\[ \int_{A} ds = 2 \left( z_b^2 + h^2 + 2Rz_b + 2Rh + 2z_bh - z_r^2 - 2Rz_r \right)^{1/2} - 2 \left( z_b^2 + 2Rz_b - z_r^2 - 2Rz_r \right)^{1/2} \] (5.5)

CASE B: $z_b < z_r < z_b + h$

\[ \int_{B} ds = 2 \left( z_b^2 + h^2 + 2Rz_b + 2Rh + 2z_bh - z_r^2 - 2Rz_r \right)^{1/2} \] (5.6)

CASE C: $z_b + h < z_r$

\[ \int_{C} ds = 0 \] (5.7)
5.2 NLC detection algorithm

The retrieval algorithm of NLC properties from SCIAMACHY is based on fully calibrated level 1 version 6.03 limb data from channel 1, more specifically radiation from clusters 3 and 4 with wavelengths between 264-300 nm. The original data are available as level 1b files and must be converted to level 1c files by the data users, determining which options to implement for the purpose of their work. For the present study, the conversion from level 1b data to level 1c was done using the scial1c tool (DLR, 2008) with the following options:

- cluster 3,4
- cal 0,1,2,4,5,6,7
- mfactordir /misc/sostrd2/mfactor/m-factor_06.01/
- type limb -ds 1,2,6,9,17,19,x0,A,B,C,D
- cat 2
- ascii-smr D0_YYYYMMDD.dat
- out filename.dat

All calibrations are performed, except for the etalon correction which should not be applied if the data is converted using the M-factor tool. Version 6.01 of the M-factor database is used. The sun mean reference spectrum (SMR) with id D0 is extracted into an ASCII file after M-factor calibration.

The level 1c file produced in this fashion is finally converted to the ASCII format by the “scia_L1C_ascii_quad” routine, so that SCIAMACHY’s vertical profiles at different wavelengths and for different geolocations can easily be read. An example of such a vertical profile of radiation scattered by the atmosphere in the absence of NLC is depicted in Figure 5.3a for three different wavelengths in the UV range. One notices that the radiance increases from 90 km down to 65 km (the so-called “knee”), where it starts to decrease monotonously with height. The altitude of the “knee” is dependent on the wavelength. This shape of the radiance profile is the consequence of a compromise between scattering and absorption in the line of sight of the instrument and corresponds to a change from an optically thick in the lower atmosphere to an optically thin regime in the mesosphere.

Figure 5.3b shows the effect of the presence of a NLC on the radiance profile. The obvious increase in radiance between 65 and 85 km is attributable to the presence of a NLC event, as expected from eq. 5.1. The positive detection of a NLC in the FoV by the retrieval algorithm indicates that either of the following criteria has been fulfilled:

1. An increasing UV radiance between 76 and 88 km at 2 wavelengths $\lambda_1$ and $\lambda_2$ is observed;

2. The radiance ratio between two consecutive tangent height measurements (between 76 and 88 km) is larger than 3 at 2 wavelengths $\lambda_1$ and $\lambda_2$ is registered.
Figure 5.3: Vertical limb radiance profile of the atmosphere for $\lambda = 265, 280$ and $295$ nm a) without and b) with a NLC in the FoV. The NLC peak in radiance occurs around 80 km, while the so-called “knee” is usually present between 50 and 70 km. The measurements were made during orbits a) 17064 and b) 12086, both in the northern hemisphere.

The choice of the wavelengths $\lambda_1$ and $\lambda_2$ singled out for the detection depends on the dimensions of the particular ground pixel examined. If the detection is carried on basically the largest limb ground pixel available, i.e., $400 \text{ km} \times 1000 \text{ km} \times 3 \text{ km}$, information from both channel 1 clusters 3 and 4 is available, i.e. any wavelength between 264 and 300 nm. In this case, the algorithm is executed for wavelengths $\lambda_1 = 265$ nm and $\lambda_2 = 291$ nm, averaged over a 2 nm boxcar. It is however possible to obtain information on a finer spatial scale for some of the measurements due to smaller integration times for cluster 4 data, i.e. for radiation with wavelength in the 283-300 nm range, leading to ground pixels which have half the across-track dimension (500 km). In this case, only data from cluster 4 are extracted and the detection is carried out for $\lambda_1 = 284$ nm and $\lambda_2 = 291$ nm, also spectrally averaged over a 2 nm boxcar. It follows that improvements of the spatial resolution come at the expense of spectral information as well as signal to noise ratio, the latter due to the shorter integration time. Figure 5.4a shows NLC limb radiance measured at 291 nm by SCIAMACHY along with the ground pixel size when using both clusters 3 and 4. The NLC limb radiance and ground pixel dimensions presented in figure 5.4b are the same measurements, but for the case where only information from cluster 4 is used. One notices that some ground pixels are about half the size of those in panel A. The dimensions of the ground pixels can be even further reduced if a special measurement campaign is taking place, requiring a SCIAMACHY Operation Change Requests (OCR). One such special campaign took place between June 30 and July 13, 2008 in order to maximize the number of measurements taken near the Arctic summer for the study of NLC. Figure 5.4c shows the smaller dimensions of the limb ground pixels which were available during that campaign.

The rate of false detection can be estimated by manually looking at a large number of single profiles and then determine how many positive detections were due to the presence of noise in the data. This can usually be identified as stochastic variations in the radiance
5.2 NLC detection algorithm

Figure 5.4: NLC radiance at 291 nm retrieved from SCIAMACHY’s nominal measurements on July 5, 2005 with different limb ground pixel size: A) 400 × 1000 km and B) 400 × 500 km. Panel C shows SCIAMACHY’s limb ground pixel size (400 × 125 km) during a special measurement campaign on July 13, 2008.

vertical profile. Using data from different years, different months and hemispheres, a false positive detection rate of about 2% is estimated. Some NLC are also too faint to be detected by the algorithm even though they might be present in the FoV during the measurement. This false negative error is dependent on the minimum conditions which must be fulfilled for the detection of a NLC by SCIAMACHY, i.e. the ratio of consecutive TH radiance measurements should be >3. This condition was set to minimize the number of false positives and is, therefore, linked to the signal to noise ratio of the instrument in this wavelength range. This limiting detection condition is in turn a function of the molecular scattering background, on top of which the NLC signal is measured, and this changes with wavelength, altitude, latitude and time.

In order to estimate the smallest detectable NLC, a reference molecular background vertical profile for different latitudes and wavelengths was computed by averaging thousands of NLC-free SCIAMACHY profiles together. All single profiles stemmed from the month of May (November), i.e. just before the start of the NH (SH) NLC season. This period is selected in order to have atmospheric conditions which are as close as possible to those during the NLC season. Figure 5.5a presents these reference profiles in their full vertical extent for different 5° latitude bands between 50° and 85° in both hemispheres and for $\lambda = 291$ nm. Figure 5.5b shows the blown-up portion of the vertical scale relevant for the NLC retrieval algorithm. Based on these profiles, the minimum NLC radiance needed to be identified as a NLC event is calculated by finding the NLC radiance that fulfills the condition of consecutive radiance ratio larger than 3. A contour plot of this minimum NLC radiance is presented in Figure 5.5c for $\lambda = 291$ nm as a function of latitude and height.

According to figure 5.5c, measurements in the SH are more sensitive to NLC, especially at
Figure 5.5: Panel A: Reference molecular background vertical profile for different latitudes and hemispheres, shown in more detail in panel B for the altitudes relevant for NLC detection. Panel C: contour plots (both hemispheres) of the minimum NLC radiance ($10^9$ photons s$^{-1}$ cm$^{-2}$ sr$^{-1}$ nm$^{-1}$) needed for an event in order to be identified as a NLC by the detection algorithm.
high latitudes, but the situation is not that straightforward. It was already shown in section 2.2 that the intensity of the scattering in the Mie regime (even for non-spherical particles) is strongly dependent on the scattering angle, considerably favoring forward scattering over backscattering. It is the case that measurements in the NH and the SH have very different scattering angles, as can be seen in figure 5.6. The scattering angles for observations in the NH polewards of 50° are in the range of 20°-75°, with the smallest values on the ascending portion of the orbit. For the SH, measurements are made in backscattering geometry with scattering angles between 115° and 155°. This means that an NLC in the NH (forward scattering) will have a very different radiance if measured in backscattering geometry, as is the case in the SH. Therefore, the radiance threshold of detection cannot easily be compared. They will be dependent on the number density and particles size. At a given latitude though, the scattering angle is the same, so SCIAMACHY’s detection algorithm is more sensitive to NLC present at higher altitudes, according to Figure 5.5.

NLC detection can be performed in both hemispheres, but the area covered in the SH is limited by the occurrence of the Southern Atlantic Anomaly (SAA). The SAA is a region where the van Allen belts of radiation, usually found at altitudes higher than 1000 km, can be found between 200 and 800 km from the surface. Since the satellite’s altitude is 799 km, it travels through this region where energetic particles trapped in the belts bombard the instrument and produce noise in the detectors. This usually drowns the optical signal and prevents the collection of useful data. Therefore, any measurements from this region are excluded from the NLC detection. The geographical limits of the SAA cover is taken to extend from 250° to 30° longitude and −60° to 0° latitude (see Figure 5.7).
5.3 Altitude

It is possible to extract information concerning the altitude of NLC peak radiance from the detection algorithm, but this information has certain shortcomings which should be highlighted. First, limb measurements usually assume that the maximum signal comes from the point tangent to the earth along the line of sight. However, the NLC scattered signal can be detected even if it is in the near- or far-field compared to the tangent point. In such cases, the registered altitude will be lower than the actual altitude of the NLC. More problematic is the limb pointing errors of SCIAMACHY, which have been characterized by von Savigny et al. (2005a) for the first 3 years of the mission. It was shown that the monthly mean tangent height errors could be as large as 2 km. For individual limb measurements, TH errors of up to 5 km were found. These values apply to an earlier version (5.04) of the SCIAMACHY Level 1 data set though. For this study, only Level 1 version 6.03 data were used that show significantly improved pointing stability and TH errors of less than \( \sim 500 \) m (von Savigny et al., 2009). Finally, SCIAMACHY’s coarse vertical resolution (\( \Delta F \)o\( V_{\text{vertical}} = 3.3 \) km) coupled with the fact that limb measurements usually occur at about the same altitudes (i.e., \( \text{TH} = 77.5, 80.8, 84.1 \) and \( 87.3 \) km) throughout the season renders single profile altitude retrieval somewhat limited in terms of information content.

Nevertheless, one can combine the retrieved single profile altitudes in daily averages which do contain useful information. A simple model was produced to determine the uncertainty of the daily mean peak altitude associated with the NLC height retrieval on a fixed altitude grid. First, a priori information on the NLC altitude (at the peak brightness) during the NLC season is used, which were taken from lidar measurements (Lübken et al., 2008) having an excellent vertical resolution. The mean altitude of the NLC is taken to be \( 83.3 \pm 1 \) km, with small variations throughout the season. No latitudinal variation is taken into account as it was deemed statistically non-significant by Lübken et al. (2008). As SCIAMACHY performs measurements on an almost fixed altitude grid, it is clear that
the uncertainty will be strongly dependent on the number of NLC detections in a day. Thus, for each day of the season and for a given number \( N \) of daily NLC detections, \( M \) ensembles of \( N \) NLC altitudes are randomly generated. This process takes into account the seasonal variation of the NLC and the natural variability, and the value of the “real” mean altitude \( \vec{h}^*(N) \) is computed for each of the \( M = 10000 \) ensembles. The next step is to simulate how SCIAMACHY would register these measurements. Here, it is simply assumed that if the modeled altitude is within the altitude bin \( TH \pm \frac{\Delta_{\text{FoV}}}{2} \), it is determined by SCIAMACHY as being at an altitude \( TH \). The mean \( \vec{h}^{\text{scia}}(N) \) of these binned ensembles of random measurements is again computed. The vector of absolute difference is constructed according to

\[
\left| \vec{h}^*(N) - \vec{h}^{\text{scia}}(N) \right| \tag{5.8}
\]

From this, the mean difference between the daily averages measured by SCIAMACHY and simulated ones can be calculated as a function of the day relative to solstice and number of measurements. The results of the simulation are presented in figure 5.8.

The uncertainty on the mean NLC altitude decreases rapidly with the number of measurements \( N \) and reaches a plateau for \( N = 50 \) of about 100 m. It should be kept in mind, however, that this simple model does not include uncertainty contribution from the near- and far-field NLC cases or from the pointing error (\( \approx 0.5 \) km).
5.4 NLC radiance

Radiance measurements of the instrument with a NLC in the FoV are composed of the molecular background and the NLC radiance. In order to obtain the NLC radiance alone, one must therefore evaluate the amount of radiance due to molecular scattering. Instead of relying on a complex radiative transfer model, the molecular background is estimated from SCIAMACHY’s limb measurements before the NLC season, as presented in figure 5.5. For every latitude band and wavelength, a reference vertical profile of molecular scattering is computed from single profiles measured throughout the first 20 days of May or November, for the NH and SH respectively. The extraction of the molecular component of the radiance at a given wavelength from a limb profile with NLC present in the FoV is done according to the following procedure:

1. Remove the stray light, i.e., the radiance measurement at the highest TH, from all measurements;
2. Shift the reference profile so that it has the same value as the limb profile at 90 km, i.e., the next-to-last TH;
3. Fit the reference background so that it coincides with the value of the limb profile at $TH_{knee}$, the tangent height just above the “knee” (see figure 5.3). The molecular background fit is computed according to the following relation:

$$I_{molecular}(\lambda, TH) = I_{REF}(\lambda, TH) + [I_{limb}(\lambda, h_{knee}) - I_{REF}(\lambda, h_{knee})] \times \left(1 + I_{limb}(\lambda, h_{knee}) - \frac{I_{limb}(\lambda, TH)}{I_{limb}(\lambda, h_{90km}) - I_{limb}(\lambda, h_{knee})}\right)$$

(5.9)

4. Remove $I_{molecular}(\lambda, TH)$ from the limb signal and calculate the brightness ratio (BR) at the NLC radiance peak for $\lambda = 291$ nm

$$BR_{291} = \frac{I_{limb}(291, h_{NLC}) - I_{molecular}(291, h_{NLC})}{I_{molecular}(291, h_{NLC})}$$

(5.10)

This procedure is an improvement over the earlier methodology which simply assumed that the Rayleigh background radiance decreased exponentially above ~70 km. As a result, the NLC radiance was calculated by subtracting a fitted exponential function between 77 and 91 km to the total SCIAMACHY measured radiance. Figure 5.9 shows 3 examples of limb profiles with a NLC in the FoV. The total radiance is shown at 3 wavelengths (265, 280, 295 nm) along with the computed molecular background for each wavelength. The rightmost panel shows the resulting NLC radiances for the 3 wavelengths.
Figure 5.9: Examples of total limb radiance at different wavelengths together with the molecular scattering fit. The rightmost panel shows the resulting NLC radiance at different wavelengths. For the sake of clarity, the radiance units ($10^{10}$ photons $s^{-1} cm^{-2} sr^{-1} nm^{-1}$) have been left out of the axis title.
The uncertainty on the NLC radiance must be calculated according to the rules of error propagation. If uncorrected stray light is not considered as well as other unquantified sources of error, it follows from eq. 5.1 that

$$L_{\lambda}^{NLC} = L_{\lambda}^{TOT} - L_{\lambda}^{Rayleigh}$$

and it follows therefore that

$$(\Delta L_{\lambda}^{NLC})^2 = (\Delta L_{\lambda}^{TOT})^2 + (\Delta L_{\lambda}^{Rayleigh})^2$$

The error on the total limb radiance, $\Delta L_{\lambda}^{TOT}$, corresponds to the spectral noise of the instrument and was estimated from dark limb measurements to be $10^7 \text{ photons s}^{-1} \text{ cm}^{-2} \text{ sr}^{-1} \text{ nm}^{-1}$ in this wavelength range. The uncertainty of the Rayleigh radiance fitted to a single limb profile, $\Delta L_{\lambda}^{Rayleigh}$, can be determined by applying the method described for the fitting of molecular scattering radiance to several (~3000) limb profiles known not to contain NLC signatures. The error then corresponds to the average absolute difference between the fitted molecular background and the actual value of the radiance. In this way, one determines a mean error which is a function of altitude, latitude and wavelength. However, it should be noted that the error is only weakly dependent on altitude and latitude and is usually found to be about $1 \times 10^9 \text{ photons s}^{-1} \text{ cm}^{-2} \text{ sr}^{-1} \text{ nm}^{-1}$. Figure 5.10 shows the dependence of the error as a function of wavelength for detections at 81 km in the 70°-75° latitude bin.
5.5 Particle Size Retrieval

Besides the NLC macroscopic attributes like occurrence frequency, radiance and altitude, the use of multi-spectral measurements enables the retrieval of microscopic properties which are especially interesting because they can provide more detailed information on the microphysics of the clouds.

Information on the size of NLC particles can be inferred from SCIAMACHY’s measurements as follows. It was shown in eq. 5.4 that the NLC radiance $L_{\lambda}^{NLC}$ is proportional to the solar irradiance, the effective number of scatterers present along the LOS and the differential scattering cross-section. The latter is in turn a function of the particle size distribution, the scattering angle and the wavelength. The equation can be rearranged in the following manner

$$\frac{L_{\lambda}^{NLC}}{S_{\lambda}} = N^* \cdot \frac{d\sigma_{scat}^*}{d\Omega}(r, \lambda, \theta)$$

(5.13)

where the left-hand side is the NLC sun-normalized radiance (also called directional albedo) while the right-hand side is the effective differential scattering cross-section of the ensemble multiplied by a factor independent of the wavelength or the scattering angle. Therefore, the NLC sun-normalized radiance is endowed with information on the NLC particle size. It can be shown that, within a limited spectral range, any effective differential scattering cross-section can be described by the following relation

$$\frac{d\sigma_{scat}^*}{d\Omega}(r, \lambda, \theta) \propto \lambda^\alpha$$

(5.14)

where $\alpha$ is the so-called Ångström exponent (Angstrom, 1929). By combining eq. 5.13 and (5.14), it follows that

$$R_{\lambda}^{NLC} = \frac{L_{\lambda}^{NLC}}{S_{\lambda}} = k \cdot \lambda^\alpha$$

(5.15)

$$\Rightarrow \ln (R_{\lambda}) = k^* + \alpha \cdot \ln (\lambda)$$

(5.16)

with $k, k^*$ being constant factors. According to eq. 5.16, the Ångström exponent can be computed by a simple linear regression with quantities involving only the NLC directional albedo and the wavelength, all available from SCIAMACHY’s measurements. This means that the Ångström exponent associated with a NLC can be experimentally determined, as will be described in more detail in section 5.5. To convert this spectral exponent to a particle size, a model of the differential scattering cross-section resulting from the ensemble of particles forming the NLC is needed. In order to do that, assumptions must be made on
Figure 5.11: Ångström exponent correction for channel 1 degradation (panel A) and for the change in solar activity (panel B).

the composition of the scatterers, the PSD and the shape of the particles. The differential scattering cross-section is modeled for different mode radii of the PSD. Based on this information, a look-up table (LUT) of the mode radius of the particles ensemble as a function of the scattering angle and the Ångström exponent in built.

Experimental determination of the Ångström exponent

All that is needed for the computation of the Ångström exponent is the NLC sun-normalized radiance for a certain number of wavelengths. The calculation of the NLC radiance having been presented in section 5.4 already, only the solar irradiance is described here. There are two alternatives for SCIAMACHY’s solar irradiance spectrum: a reference irradiance spectrum or the daily mean solar spectrum.

Early determination of SCIAMACHY NLC particle size (von Savigny et al., 2004) was done using a SCIAMACHY reference spectrum from Skupin et al. (2003). That solar spectrum is assumed to have smaller uncertainties than the daily averaged solar irradiance due to manual correction and comparison with solar irradiance measurements from other instruments. However, as the reference spectrum is constant in time while the transmission of channel 1 decreases with each passing year (see figure 4.5) - a change which is wavelength dependent - SCIAMACHY limb radiance without the appropriate corrections and normalized with such a solar irradiance spectrum exhibits spurious spectral features which can greatly affect the value of the measured Ångström exponent and hence the determined NLC particle size (von Savigny et al., 2007c). Figure 5.11a shows the value by which the measured Ångström exponent is biased as a function of time if the reference solar spectrum is used in combination with limb radiances not corrected for channel 1 degradation.

Another effect arising from the use of a fixed solar reference spectrum for the radiance
normalization is that one overlooks the natural variation of the solar spectral features during the 11-year solar cycle. Depending on the solar activity, the irradiance of our star changes slightly, and this modulation is spectrally dependent, inducing another artificial contribution to the measured Ångström exponent, as shown in figure 5.11b. The impact of this artifact on the spectral exponent is however very small, especially with regard to the error induced by overlooking channel 1 degradation: the quantitative change of $\alpha$ is of the order of 0.02 for the 27-day solar cycle and 0.07 for the 11-year solar cycle while it is found to be between $-0.2$ and $1.5$ for the degradation of channel 1 (Robert et al., 2009).

Due to these problems linked with the reference spectrum, the daily averaged solar irradiance measured by SCIAMACHY is used to normalize the NLC radiance. This has the benefits of being insensitive to the degradation correction, as both the radiance and daily average solar irradiance are corrected in the same way, and its variation during the course of the solar cycle is taken into account. The relative uncertainty regarding the daily mean solar irradiance in the 265-300 nm range is of the order of 10%, as can be observed in figure 5.12 taken from Skupin et al. (2003). This relative error is estimated from the difference between SCIAMACHY’s solar irradiance and that of the SOLSTICE and SUSIM instruments.

The uncertainty of the calculated NLC sun-normalized radiance $R_{\lambda}^{NLC}$,

$$R_{\lambda}^{NLC} = \frac{1}{S} \cdot \left( L^{TOT} - L^{Rayleigh} \right)$$ (5.17)

stems from the error on the total radiance measurement, the uncertainty of the Rayleigh background modeling and that of SCIAMACHY’s daily mean solar irradiance. According to error propagation principles, it is found to be
where the explicit indication of the variables dependence on the wavelength has been omitted for clarity.

As the NLC sun-normalized radiance is now well characterized, we can turn our attention to the determination of the spectral exponent through linear fitting, as mathematically expressed by eq. 5.16. It should be noted that all emission features and strong Fraunhofer lines in the 265-300 nm range, shown in figure 5.13, are excluded from the fitted spectral window as they might affect the resulting \( \alpha \) determination.

Figure 5.14 shows two examples of a fit of the logarithm of SCIAMACHY’S directional albedo as a function of the logarithm of the wavelength, for cases with different degrees of scatter. The \( \alpha \) exponent calculated from the linear regression is also indicated in the plots as well as the error associated with it. The uncertainty regarding the spectral exponent is calculated from the least-square fitting routine.

Although it is the case for all regressions presented in figure 5.14, the uncertainty on the spectral exponent \( \Delta \alpha \) is not always \( \approx 0.15 \). Further investigations indicate that the error is a function of the NLC directional albedo and, to a lesser degree, the NLC altitude. Figure 5.15a illustrates the dependence of the spectral exponent uncertainty \( \Delta \alpha \) on the directional albedo at \( \lambda = 291 \) nm and for 2 NLC altitudes. The error on the Ångström exponent is smaller for higher NLC and very bright clouds, and goes asymptotically to about 0.14. Figure 5.14b and c show the relative contribution of the solar irradiance and Rayleigh background radiance uncertainty, respectively, to the total uncertainty of the spectral exponent. For bright clouds, the limiting factor determining the error on the spectral exponent is the uncertainty related
5.5.1 From Ångström exponent to particle size

The knowledge of the Ångström exponent of a NLC event at a given scattering angle can in principle provide information on the NLC particle size, but one must make assumptions concerning the PSD and the shape of the particles. Once reasonable assumptions are made, the T-matrix method can be used to compute the differential scattering cross-section of an ensemble of spherical and non-spherical particles. Not all particle shapes can be modeled by the T-matrix approach, but oblate and prolate spheroids as well as cylindrical particles can be adequately modeled. If non-spherical particles are assumed, one must also conjecture on the orientation of the particles, but it is usually assumed that the particles are...
randomly oriented. These models can finally be translated into look-up tables, containing the Ångström exponent for different scattering angles and PSD mode radii which are used to convert experimental Ångström exponents into NLC particle size.

An important aspect of the modeling concerns the composition of the NLC particles. There is now considerable evidence that NLC particles consist mostly of water (Hervig et al., 2001). The critical physical property to the modeling of scatterers is the refractive index, $\eta_{\text{ice}}$. For this work, the complex refractive index of water ice from Warren (1984) and Warren and Brandt (2008) is used. Both the real and imaginary part are presented in figure 5.16. The imaginary part of the ice refractive index exhibits a deep minimum between 200-400 nm so that no or negligible absorption takes place in this spectral range. The real refractive index changes slightly in the 265-300 nm range, from 1.35 to 1.33, and even though this variation seems inconsequential, it must be taken into account when fitting the spectral exponent. Earlier work regarding SCIAMACHY particle size retrievals used the mean ice refractive index in this range when modeling the scattering properties of an ensemble of ice particles. Nevertheless, it became clear after implementing the complete information of $\eta_{\text{ice}}$ in the retrieval that even such small variations - of the order of 1% of the average value - affect our results significantly. The use of $\eta_{\text{ice}}$ tends to increase the value of the particle size, the magnitude of the increase being strongly dependent on the assumed PSD. For a normal PSD with $\sigma = 22$ nm, the increase is found to be between 10 and 20 nm.

For this work, the LUT calculated for ensembles of water ice particles have been provided by Gerd Baumgarten who used a modified version of the T-Matrix code from Mischenko and Travis (1998). With these tables, it is possible to retrieve NLC particle sizes for the following assumptions:

- Normal PSD with $\sigma$ in the 10-30 nm range, all shapes
5.5 Particle Size Retrieval

- Lognormal PSD with $\sigma$ in the 1-1.6 range, all shapes
- Spheroids with axial ratio in the 0.2-5 range, randomly oriented
- Cylinders with axial ratio in the 0.2-5 range, randomly oriented

Figure 5.17 shows such tables for a variety of PSD and shapes. The figures show a contour plot of the Ångström exponent as a function of the scattering angle and the mode radius of the PSD. For a given Ångström exponent measurement $\alpha$ made at a scattering angle $\theta$, one follows the vertical line corresponding to $\theta$ on the plot to find the value of $r$ for which the LUT $\alpha$ is the same as the experimentally determined one. It was already mentioned that SCIAMACHY's measurements are done at different scattering angles depending on the hemisphere and for the portion of the LUT relevant for the SH ($115^\circ$ and $155^\circ$). We notice that a problem arises for some of these look-up tables because a given Ångström exponent can correspond to more than one mode radius in the 0-150 nm range, assumed to be the confines within which NLC particle sizes exist. For instance, if it is assumed that an NLC particle population can be described as spheres with a Gaussian PSD characterized by $\sigma = 22$ nm and that the measured spectral exponent is -1.0 for a scattering angle of $140^\circ$, the retrieved particle size could be simultaneously 25, 73 and 98 nm as can be seen in figure 5.17. Thus, the size of NLC particles in the SH cannot be determined unambiguously and no rigorous retrieval is feasible in this region. In the NH however, scattering angles are in the $20^\circ$-$75^\circ$ range and, according to the different LUT, this region does not pose any difficulty for the retrieval of the NLC particle size. There is, nonetheless, a drawback when determining the particle size at small scattering angles. For a given error on the Ångström exponent $\Delta \alpha$, the uncertainty of the corresponding radius will be much larger due to the smaller sensitivity of the Ångström exponent on the mode radius. The particle size retrieved in the ascending portion of the NH orbit will consequently have larger uncertainties in general.
5 Retrieval algorithms

Figure 5.17: Contour plots of the Ångström exponent as a function of the scattering angle and the PSD mode radius for ensembles of NLC particles with different shapes (spheres, prolate and oblate spheroids, cylinders) and PSD (Normal, $\sigma = 22$ nm and Lognormal, $\sigma = 1.4$).
NLC properties part I: Occurrence, radiance and altitude

The previous chapter described in detail how SCIAMACHY’s spectral measurements can be used to retrieve NLC properties from space. The results obtained from applying the detection algorithm to all available data from SCIAMACHY (2002-2009) are presented in this chapter. The focus lies on the description of NLC macroscopic properties from SCIAMACHY retrieved over seven years, namely occurrence frequency, radiance and altitude. A large part of the results presented in this chapter were published in Robert et al. (2009). Measurements of NLC particle size will be surveyed in the next chapter. The spatial and temporal distribution of NLC properties will also be presented and discussed for both hemispheres. This will include an analysis of the short- and long-term variations of the properties and the physical causes underlying these variations. The SCIAMACHY climatology will also be compared with model results and measurements from other instruments including other NLC data sets as well as measurements of water vapor and temperature from MLS.

6.1 NLC Occurrence

More than 67 000 NLC were detected by SCIAMACHY since the beginning of its operation in August 2002. The number of detections alone is, however, not representative of the NLC activity over the season since the geographical coverage can change from day to day and year to year depending on the instrument’s operations. Instead, the daily occurrence frequency is employed as an indicator of NLC activity. It is computed as the ratio of the
number of limb measurements with an NLC detected in the FOV, for a given latitude band and day, to the total number of limb measurements in that same latitude band and day. Except when stated explicitly, the occurrence frequency usually assumes zonal symmetry and therefore longitude is not taken into account for its calculation.

Figure 6.1 presents the daily NLC occurrence rate as measured by SCIAMACHY for three different latitude bands (55-65°, 65-75° and 75-83°) and for both hemispheres. The data show the daily NLC occurrence rate averaged over years 2002-2009. The plots display the main features of a typical NLC season.

In the NH, the season starts around day −30 relative to the summer solstice (DTS). There is a small incidence of clouds at the beginning of the season, but as the upper mesosphere temperature decreases, the frequency of occurrence increases rapidly until it reaches a plateau near 0 DTS. From that point on, the occurrence rate increases only slightly until it reaches a maximum, typically between 10 and 40 DTS. Finally, the occurrence of NLC decreases until the end of the season, which is around 70 DTS. The behavior of the occurrence rate at different latitudes generally follows this pattern, although the maximum occurrence frequency reached differs considerably, increasing with latitude and reaching typically 100% of cloud coverage during the core of the season at latitudes larger than 80°.

The SME climatology from Olivero and Thomas (1986) depicts a similar evolution of NLC occurrence over the summer season, with a peak in NLC activity around 20 DTS and more frequent NLC detection at latitudes polewards of 70-75°. Also, the start of the NH season is in good agreement with early results from the Solar Occultation For Ice Experiment (SOFIE) instrument, which is more sensitive to the presence of icy particles than SCIAMACHY (Hervig et al., 2009), and observes a commencement of the season at -31 DTS for the summer season 2007 at 70°.
Interhemispheric differences

The seasonal pattern in the SH is similar to that of the NH. The first clouds visible to SCIAMACHY form around −30 DTS and the occurrence frequency increases rapidly until about 10 DTS. The clouds finally vanish at the end of the summer, usually some time around 60 DTS. Similarly to the NH, there are less NLC detected at lower latitudes. This is especially evident when the season’s occurrence rate maxima at 75-83° and 65-75° are compared, which differ by a factor of nearly 2. According to figure 6.1, there is no distinct plateau reached for the NLC occurrence rate in the SH, as is the case for the NH. Instead, considerable variability can be observed, even during the core of the season. This is attributed to the generally higher temperatures (4-7 K) in the SH summer mesosphere near 80 km (Hervig and Siskind, 2006; Wrotny and Russell, 2006) compared to the NH. Also, the fact that the NH winter stratosphere is less stable than the SH winter stratosphere makes the SH NLC season more prone to variability, an effect attributable to the interhemispheric coupling, as described by Karlsson et al. (2007).

There are also fewer clouds detected in the SH relative to the NH. However, caution must be exerted when comparing the interhemispheric data, since the scattering angles are much larger in the SH, as was already discussed in section 5.2. As the amount of UV radiation scattered by nanometer-size particles is considerably dependent on the scattering angle, NLC in the SH scatter less radiation. This makes them appear dimmer than their NH counterpart and therefore more difficult to detect. Comparing both hemispheres would require a knowledge of the PSD and the particle shape. It can therefore not be unambiguously stated from the current data set that there are less NLC in the SH than in the NH. Different instruments and models (Hervig and Siskind, 2006; Wrotny and Russell, 2006; Bailey et al., 2007; Lübken and Berger, 2007), however, support this conclusion. The Student Nitric Oxide Experiment (SNOE) (Solomon et al., 1996), for instance, measured NLC in limb geometry in both hemispheres between 1998-2003 (Bailey et al., 2005), but with small (∼5°−55°) scattering angles for SH observations and larger (∼120°−175°) scattering angles in the NH. Despite the advantageous forward scattering geometry in the SH, more NLC were detected in the NH. Further analysis of SNOE data (Bailey et al., 2007) gathered during a special measurement mode with similar observing geometry in both hemispheres (scattering angle of ∼125°−150°) confirmed the asymmetry. The results obtained by this investigation showed that for the period 0-40 DTS, the ratios of occurrence frequency in both hemispheres (OF\textsubscript{NH}/OF\textsubscript{SH}) were 1.28 at 80°, 2.43 at 70°, and 4 at 60°. This is quite similar to SCIAMACHY’s observations for which the interhemispheric occurrence rate ratios averaged between 2002-2009 are 1.20 at 80°, 2.3 at 70°, and 2.7 at 60°. The fact that the ratio increases with decreasing latitude is in very good agreement with the Leibniz Institute Middle Atmosphere (LIMA) model (Berger, 2008) which shows an increase in the temperature difference between the hemispheres for decreasing latitudes (Lübken and Berger, 2007). As to the small discrepancies between the ratios from SNOE and SCIAMACHY, they cannot be explained solely on the grounds of the difference in observing geometry since the SNOE data show larger ratios than the SCIAMACHY data, even though the contrary
Figure 6.2: Contour plots of the mean supersaturation during the summer season near the mesopause derived from MLS temperature and water vapor measurements between 2004-2009. The variation of the saturation ratio with time and latitude is presented for both hemispheres at different altitudes (82, 84, 86 and 88 km).
would normally be expected due to SCIAMACHY’s increased sensitivity to NH NLC. Local
time does not play a role either, as the measurements were done around 10 AM with both
instruments. It is important to keep in mind however that the analysis from (Bailey et al.,
2007) is based in single seasons whereas the results from SCIAMACHY are calculated for
the mean of years 2002-2009. Although a more complete study should be done in order to
come to some reliable conclusions, it seems that despite the large difference in scattering
angles in both hemispheres, SCIAMACHY samples a large portion of the NLC population in
the SH.

The onset of the NLC season in both hemispheres occurs at about the same time, yet
the southern hemispheric NLC season appears to be about 10 days shorter than that of the
NH. This observation is in disagreement with several studies which concluded that both
seasons are of equal length, but that the southern hemispheric NLC season starts 10-15 days
early than in the NH (Wrotny and Russell, 2006; Bailey et al., 2007). However, the shorter
NLC season in the SH as observed by SCIAMACHY may be partly be caused by the lower
detection sensitivity in this hemisphere, especially at the beginning of the season when
clouds are dimmer.

Saturation of the mesopause

The characteristics of the NLC season can be largely explained by the saturation ratios found
near the mesopause. Figure 6.2 shows, for both hemispheres, the mean supersaturation
for different altitudes as a function of time and latitude. These saturation ratios were
derived from MLS measurements of temperature and water vapor between 2004-2009,
using the expression from Marti and Mauersberger (1993) to calculate the ice vapor pressure.
The levels of supersaturation are higher at higher altitudes, where ice particles form and
grow while sedimenting to lower altitudes. As will be presented in section 6.3, the peak
radiance of NLC in the NH is usually found between 82 and 84 km. The supersaturation
at 84 km shows a variation which is very similar to the occurrence rate, with a saturation
ratio of 1 occurring for the first time around −28 DTS and ending 67 DTS, being largest
between −10 and 40 DTS and extending to lower latitudes, down to about 55°. This is
in general agreement with the SCIAMACHY results for the NH, even though some (albeit
few) NLC are also detected at latitudes lower than 55°. Small-scale perturbations can
easily drive the atmosphere into a state of supersaturation (or out of it), which could
explain the occasional detection of NLC at lower latitudes. The fact that the atmosphere
is predominantly supersaturated throughout the entire summer season would imply that
ice particles should be present most of the time. SOFIE measurements do concur with this
statement, by showing that for the 66-80° latitude band, ice is present more than 80% of
the time between −25 DTS and 65 DTS (Hervig et al., 2009). The supersaturation in the
SH is generally lower than in the NH at a given altitude, explaining the lower frequency
of occurrence observed. In the SH, the NLC are usually detected somewhere between 83
and 86 km. The map of saturation ratios at 86 km shows that at high latitudes, the season
Figure 6.3: Daily NLC occurrence rate for latitudes higher than 50° in the northern hemisphere for seasons 2002-2009. The occurrence rate was calculated for latitudinal bins of 5°. The lower right panel shows a contour plot of the mean meridional and temporal variation NLC occurrence frequency for the NH summer.

should last between −30 DTS and 60 DTS. This is also in quantitative agreement with the results from SCIAMACHY regarding the beginning and end of the season.
Individual seasons

Figure 6.3 reveals the spatial and temporal variation of NLC occurrence frequencies for the individual years 2002-2009 in the NH, as well as a contour plot of the average of seasons 2002-2009 in the lower right panel. A 5-day running mean has been applied to the data. The latitudinal dependence of NLC occurrence is unmistakable in such a representation. The season is roughly symmetric around its maximum, except maybe for the early end of the season shorter for latitudes lower than \( \sim 65^\circ \). The occurrence rate reaches values close or equal to 1 at high latitudes during the core of the season while less NLC will be observed at lower latitudes. Besides the general seasonal features which were presented earlier, we can observe variations on shorter timescales which can be the consequence of a variety of atmospheric processes such as gravity waves, planetary waves, tides and interhemispheric coupling.

Year-to-year variations are also observable. Season 2004 for instance features an early onset of NLC at low latitudes compared to a typical season, while 2003 exhibits more abrupt variations of the occurrence rate over the season. The length of the season can also vary, as can be seen for years 2003 and 2005 where the seasons were shorter than 2004 and 2007. It is important to keep these variations in mind as they can have a large impact on a yearly mean which will be presented in section 6.4.

The NLC occurrence rates in the SH for all seasons observed by SCIAMACHY are presented in figure 6.4. The lower right panel of figure 6.4 shows the mean NLC occurrence rate contour averaged for years 2002-2009 SH. There are distinctly less NLC detected in the SH as compared to the NH. Relatively few NLC are observed at latitudes lower than 60\(^\circ\), and if so, they are hardly ever observed 2 days in a row. This fact is however not discernible from the plots as a 5-day running mean has been applied to the data for a clearer representation of the seasonal variation. There are also generally much more variations over the season and an activity peak earlier in the season compared to the NH. The second half of the season is noticeably less active. Investigations by Morris et al. (2009) using ground-based radar and MLS temperatures in the SH have shown that the usually weaker January and February NLC (and PMSE) activity is linked to an enhancement in westward propagating quasi 2-day planetary waves, probably originating from the low-latitude easterly jet. This feature of the SH summer is recurrent from year to year, but varies in strength. The 2-day waves are also observed in the NH, but they are weaker there. Since the SH summer mesopause is warmer than the NH summer mesopause, the heating produced by such wave activity affects the SH ice layer to a greater extent.

Solar proton events

A sudden change in NLC occurrence rates is observed during the SH season 2004/2005 at 32 DTS, corresponding to January 16, 2005. This depletion of NLC was identified by von
Figure 6.4: Daily NLC occurrence rate for latitudes between 50°-83° in the southern hemisphere for seasons 2002-2009. The occurrence rate was calculated for latitudinal bins of 5°. The lower right panel shows a contour plot of the mean meridional and temporal variation NLC occurrence frequency for the SH summer.
Savigny et al. (2007b) as a possible consequence of a solar proton event (SPE). Figure 6.5 shows in more detail the variation of the NLC occurrence rate at 70° and 80° during the SPE, as well as the temperature response at 86 km. An increase in temperature of about 10 K was observed over 3 days after the SPE at high latitudes, while it took approximately 5 days at lower latitudes for the maximum change in temperature to take place. Although the high latitude NLC could recover after the event, the occurrence rate at 70° dropped from 80% to ~30%, not to increase again afterwards. Mechanisms proposed to explain the sudden disappearance of NLC were Joule heating in the mesopause region, direct particle heating due to collisions of the high-energy particles and catalytic destruction of O3 (von Savigny et al., 2007b). In order to further investigate the possible mechanisms responsible for the NLC disappearance during an SPE, Becker and von Savigny (2009) modeled the effect that a reduced diabatic heating in the middle mesosphere due to ozone destruction would have on temperatures near the summer mesopause. The associated reduction in solar heating is shown to affect zonal wind and the filtering of gravity waves, which leads to a reduced residual circulation causing reduced adiabatic cooling at polar summer mesopause. The temperature changes caused by these physical processes can explain the sudden decrease in NLC occurrence frequency and support the proposed mechanism.

Zonal dependence of NLC occurrence rates

Although it is customary to present and calculate zonally averaged NLC occurrence frequency, some information can also be gained by studying the variation of NLC properties in different longitude bins and in a given latitude band. One phenomenon which can be observed by looking at non-zonally averaged occurrence rate is, for instance, the quasi 5-day planetary wave known to be present at the summer mesopause (Kirkwood and Stebel, 2003; Merkel et al., 2003).
Figure 6.6: Panel A: NLC occurrence rate averaged between 60°-80° latitude as a function of longitude and day relative to solstice for NH summer season 2005, accompanied by its wavelet power spectrum. Panel B: Same as panel A but for MLS temperature at 85 km.

An examination of the NLC data set from SCIAMACHY shows that the 5-day wave signature can be identified quite clearly, especially at the beginning of the season (von Savigny et al., 2007a). Figure 6.6 displays the variation of the NLC occurrence frequency as a function of longitude and time for the NH summer season 2005 (panel A) as well as the temperature anomaly fluctuations at 85 km (panel B) for the same period. The figure also presents the wavelet power spectrum of the occurrence rate and the temperature over the season. Both of these variables show a clear westward propagating 5-day wave of wavenumber 1, i.e., the zonal variation on a given day corresponds to a $2\pi$ cycle of a sine wave. The wavelet power spectrum shows that this type of wave is especially active at the beginning and end of the NLC season, both with regard to temperature and NLC occurrence frequency. The temperature is in opposite phase with the NLC occurrence rate, as expected, and has an amplitude of approximately 3 K. The excellent correspondence of the minimum in temperature and the maximum in NLC occurrence frequency suggests that the quasi 5-day wave signature in the temperature field is the main driver for change witnessed in NLC occurrence. The 5-day planetary waves are thought to be excited by the moist convection in the troposphere (Miyoshi and Hirooka, 1999), in-situ baroclinic instabilities of the summertime jet near the summer mesopause (Garcia et al., 2005) and gravity waves (Lawrence and Jarvis, 2003). Investigations by Jarvis (2006) have established that the 5-day wave activity is modulated by the 22-year Hale cycle, showing a connection with solar activity.

Another process which can be observed in the longitudinal dependence of NLC occurrence
rates is the forcing by gravity waves. Although Thomas and Olivero (1989) did not find significant differences in NLC activity at different longitudes in the SME data set, recent results from Chandran et al. (2009b) provide evidence of zonal differences in NLC occurrence frequency due to gravity waves. A study of SCIAMACHY NLC occurrence frequency separated in 12 longitude bins and averaged between 60°-80° latitude reveals marked differences in the general behavior of the occurrence frequency for different longitudes in the NH. Figure 6.7 displays contour plots of the occurrence frequency for all NH seasons and different longitudes. Planetary wave signatures, such as those illustrated in figure 6.6, are not discernible in this figure because a 20-day running mean was applied to the data in order to observe only the general behavior of the occurrence rate over the season. Despite the identical coverage of observations during the season, adding up to about 9-12 detections per longitude bin per day, a recurrent pattern of reduced NLC activity can be seen throughout most years between 270°-330° and, to a lesser extent, between 90°-150°. A region of elevated NLC activity is also observed between roughly 0°-60°.

Although a more thorough investigation of this effect is needed, these early results are in excellent agreement with the analysis by Chandran et al. (2009b). Using data from the Cloud Imaging and Particle Size (CIPS) experiment, they were able to detect gravity waves within the NLC layer during the summer season (Chandran et al., 2009a) and to identify regions of enhanced gravity wave activity. Results from the analysis of the 2007 NH season are presented in figure 6.8. Longitude bands corresponding to increased gravity wave activity are in the range of 60°-180° and 260°-330°. Longitude bands corresponding to dampened gravity wave activity are found between 340°-80° and 180°-220°. These results concur with SCIAMACHY’s observations of NLC activity over the season for most years. Chandran et al. (2009b) found that regions of enhanced gravity wave activity were highly correlated with temperature changes (of the order of 2 K) and anti-correlated with NLC occurrence frequency. This suggests that these waves contribute to a local warming of the atmosphere and a decrease of NLC occurrence frequency. The investigation by Chandran et al. (2009b) also shows that the amplitude of the forcing seems to change from year to year, while regions of lowest wave activity and highest PMC occurrence remain the same. Variations of the occurrence rate with longitude in SCIAMACHY’s data set are, however, only observed in the NH summer season, while CIPS data show that a similar signature is also present in the SH. This might be linked to the reduced sensitivity of SCIAMACHY to the SH ice layer already discussed previously.

6.2 NLC radiance

Although the occurrence rate is a good indicator of NLC activity and is simple to retrieve, it suffers from a limited dynamic range and does not represent a real physical quantity. The NLC radiance (or similarly the directional albedo) improves on these shortcomings by revealing not only whether or not clouds are present, but also how strong they can
Figure 6.7: Zonal dependence of the NLC occurrence rate during the NH summer season for years 2003-2009. Note the recurrent decrease in NLC occurrence rate for longitudes between 270°-330° and, to a lesser extent, between 90°-150°.
scatter light. The radiance is a function of the particle size and the number of scattering particles in the ice layer, and can vary over orders of magnitude. However, it also entails some disadvantages of its own. The main difficulty in interpreting the NLC radiance from satellite measurements lies in its strong dependence on the scattering angle during the observation. A space-borne instrument in a sun-synchronous orbit, such as SCIAMACHY, scans the atmosphere in such a way that the scattering angle is nowhere the same along the orbit. This complicates the measurements made at different scattering angles.

Figure 6.9 shows the time series of all NLC radiance observations at \( \lambda = 291 \) nm from SCIAMACHY. It is clear that the SH radiance is much smaller than that for the NH. This results most probably from the large scattering angles in the SH and, to a lesser extent,
from the weaker NLC activity there. However, since the daily satellite coverage is mostly the same in terms of latitudes (and hence scattering angles), the daily averaged radiance can be compared within a season. Moreover, radiances measured at about the same latitude (either in the ascending or descending part of the orbit) can be compared with one another since they are observed at similar scattering angles.

The mean daily NLC radiance at $\lambda = 291$ nm for different latitudes in both hemispheres calculated from SCIAMACHY data between 2002-2009 is shown in figure 6.10. The seasonal features are similar to those of the occurrence frequency, with larger radiances at higher latitudes. One interesting difference is the lack of a constant plateau reached after a certain period, as was the case especially in the NH at latitudes larger than 65°. Instead, the NH radiance increases from the beginning of the season until about 30 DTS, when it rather suddenly decreases. The NLC radiance at high latitudes shows much more variation than the occurrence frequency. This proves to be a very interesting attribute when trying to quantify variability of NH NLC activity polewards of 75°. The seasonal variation of the SH NLC radiance however does not differ notably from the occurrence rate climatology. It increases until about 10-20 DTS and then weakens towards the end of the season, more abruptly and later at high latitudes, and more gradually for latitudes equatorwards of 75°. The variation in NLC radiance is, however, more structured than in occurrence frequency.

The daily NH NLC radiance at $\lambda = 291$ nm for each year between 2002-2009 and latitudes polewards of 50° is presented in figure 6.11. Note that the color in the contour plots changes according to a logarithmic scale which varies between $1 \times 10^{9}$ and $1 \times 10^{12}$ photons s$^{-1}$ cm$^{-2}$ sr$^{-1}$ nm$^{-1}$, i.e., a variation over three orders of magnitude. The average seasonal variation for all years is also presented in detail in the lower right panel of the figure. The seasons share similarities with the occurrence frequency, except that the
scale of the variations over the season is much larger than that for the occurrence rate.

The daily NLC radiance in the SH for all available years of SCIAMACHY and different latitude bins between 50 and 83° is presented in figure 6.12, similarly to figure 6.11. The color scale is the same as for the previous figure and, once more, definitively lower values of the NLC radiance can be observed in the SH. The mean SH NLC radiance seems to be more symmetric than in the NH relatively to the peak of the season, but it is also possible that the NH values are biased by the very strong NLC activity which took place between 20-40 DTS...
6.3 NLC altitude

The NLC peak radiance altitude observed by satellite instruments is difficult to interpret because, as noted by Thomas (1984), the true altitude of the layer is not necessarily the
observed tangent height of the maximum radiance. It is therefore especially interesting to look at the results obtained from SCIAMACHY and compare them with other measurements to see how well they agree. It is important to keep in mind the main limitation of the altitude retrieval by SCIAMACHY, already pointed out in section 5.3. As the instrument performs limb measurements on an almost fixed altitude grid (roughly 78, 81, 84 and 87 km), the uncertainty is largely dependent on the number of measurements made in a day and on the altitude of possible cloud observations.

Just as for the occurrence rate and the radiance, the averaged seasonal variation of NLC altitude is presented for both hemispheres and different latitude bands in figure 6.13. The grey area surrounding the different curves represents the error of the mean and amounts to about ±200 – 300 m, depending on the number of NLC detected and, hence, the latitude. In the NH, the NLC are first detected at altitudes near 84 km, but the peak radiance subsequently goes down by about 2 km in 50 days to reach heights slightly lower than 82 km. Past 30 DTS, the peak altitude moves up in the mesosphere to return to ∼ 84 km at the end of the season. During the core of the NH season (from 0 to 40 DTS), the NLC peak radiance altitude is apparently dependent on the latitude band, with the lowest clouds detected at the highest latitudes. The amplitudes of the difference in altitudes are however similar to the error of the mean. In the SH, the NLC altitude is generally 1-1.5 km higher than in the NH, starting between 84 and 85 km at −30 DTS and descending to about 83.5 km at 30 DTS. From then on, the NLC peak brightness altitude increases slightly to reach 84 km at the end of the season, around 60 DTS. There is no notable difference in NLC altitudes for different southern latitudes, except at the beginning of the season, where at high latitudes NLC occur at great heights than their low latitude counterparts. Variability of NLC altitude during this early period is, however, largely due to the relatively small number of NLC measurements.

The variation of NLC altitude over the season is a feature already observed by different instruments and models. Figure 6.14a shows the daily mean NLC altitudes obtained from the LIMA model for the NH 2001 and SH 2003/2004 seasons. There is good qualitative agreement between SCIAMACHY and LIMA daily NLC altitudes in the NH, with the lowest NLC found between 10 and 40 DTS. The SCIAMACHY altitudes are however 0.5-1 km lower than those in the model. Latitudinal variation of the altitude on the order of 40m/deg at low latitudes (and more gradual slope at higher latitudes) is also found in the model (Lübken and Berger, 2007), even though it is not apparent in figure 6.14a. In the SH, the model results are also generally consistent with the SCIAMACHY results. The NLC are found at altitudes near 85 km at the beginning of the season and go down to heights between 83.0-83.5 km at 20 DTS. The SCIAMACHY minimum altitude is found later at around 30 DTS, but the LIMA model is representative of just one season whereas the SCIAMACHY data set is averaged over seasons 2002-2009, which might explain the discrepancy.

Figure 6.14b presents the daily NLC altitude averaged over seasons 2003-2006 and for the 65° – 75° latitude band (Kristell Pérot, personal communication, 2009) obtained with the Global Ozone Monitoring by Occultation of Stars (GOMOS) instrument (Kyrölä et al.,
2004). This instrument has an improved altitude accuracy over SCIAMACHY with a vertical resolution better than 1 km. Results in the NH show NLC altitudes of about 84 km at the beginning and end of the season, as well as lower NLC during the core of the season, reaching a minimum altitude of about 82 km at 20 DTS. The results are similar to those of SCIAMACHY, except that the minimum altitude registered by SCIAMACHY is lower by about 0.3 km. In the SH, SCIAMACHY’s daily NLC altitudes are generally higher than those measured by GOMOS by about 0.5-1 km, but the asymmetry of the seasonal variation of both instruments is very similar, with a minimum NLC altitude around 30 DTS. Interestingly, GOMOS also observes the local maximum at around -20 DTS.

The altitude of the NLC layer is linked with the altitude at which a transition from saturation to subsaturation occurs in the upper mesosphere. As explained by Chu and Gardner (2003), icy particles forming near the mesopause slowly settle through the saturated region due to the action of gravity and grow by direct deposition of water vapor on their surfaces. Upwelling air masses from below, resulting from the consequence of the meridional circulation, also act upon the particles in such a way that they experience buoyancy. This extends the period of time during which the particles are found in a saturated environment. Finally, as the particles reach the subsaturated atmosphere, they quickly sublimate within a few hundred meters distance (Witt, 1961). In order to verify whether this explanation is supported by SCIAMACHY’s measurements, contour plots of the saturation ratios in the upper mesosphere as a function of height and time and for different latitude bands are presented in figure 6.15, along with the daily mean NLC altitude from SCIAMACHY. The saturation ratios are derived from MLS data. The seasons presented, NH 2006 and SH 2004/2005, are representative of all years. In the NH, the NLC daily mean altitude follows very closely the transition region for which $S \approx 1$, with differences of only a few hundred
meters. The agreement is especially good at higher latitudes. In the SH, there is a larger disparity between the $S = 1$ isoline and the daily mean NLC altitude, more particularly during the core of the season. SCIAMACHY overestimates the NLC altitude by almost 1 km, a result similar to the comparison of SCIAMACHY SH NLC altitudes with GOMOS and LIMA. Nonetheless, some of the variability of the $S = 1$ isoline is mirrored in the NLC altitude variation, mostly at high latitudes.

The limited agreement of the SH NLC altitude with other data sets, while the NLC altitudes derived in the NH seem to concur with them, possibly arises from the fact that the mean SH NLC height is situated near 84 km. This situation makes it much less likely that SCIAMACHY detects SH NLC at other heights than at the 84 km TH due to its fixed vertical grid whereas in the NH, since the mean height is usually lower (82-83 km), the detection takes place at both the 81 and 84 km TH, and in this fashion a representative mean NLC altitude can be calculated. The most unexpected conclusion which is reached from these observations is not that SCIAMACHY does not resolve well the SH daily NLC heights, but rather that the NH daily NLC altitudes are in good agreement with other measurements despite the coarse
Figure 6.15: Vertical profiles of the saturation ratio in the upper mesosphere throughout the NH 2006 (left panel) and SH 2004/2005 (right panel) season, along with the SCIAMACHY daily mean NLC altitude (dashed). The profiles are shown for various latitudes. The solid grey line surrounding the dashed curves represents the error of the mean daily NLC altitude.
6.4 Time series of NLC properties

The previous sections presented NLC properties and their variation mostly on a seasonal timescale. If phenomena such as planetary waves and solar proton events can be studied by examining day-to-day variation of NLC properties, secular trends and solar cycle effects are the focus of the investigation when studying inter-seasonal variation of NLC. In the case of SCIAMACHY, only seven full NLC seasons are available in each hemisphere. On the basis of such a limited time series, the determination of trends is not possible. This is especially true when considering the much larger effect of the 11-year solar cycle on NLC properties compared to the possible trend derived from SBUV data by DeLand et al. (2007) and Shettle et al. (2009). Since SCIAMACHY started its measurements in late summer 2002, an appreciable part of the descending solar cycle 23 has been sampled, as can be observed in figure 6.16, and it is hence conceivable that a solar signature in the year-to-year variations of NLC properties could be detected.

Figure 6.17a shows the evolution of the seasonally averaged NLC occurrence frequency for different latitude bands and for both hemispheres. In order to keep the figures clear, no error bars representing the seasonal variability are shown; they are instead presented in table 6.1. The NH season 2002 is not shown since only the last three weeks of the season were observed and the result would undoubtedly be biased. First and foremost, no clear trend is discernible in either the NH or the SH. The year-to-year variability in the NH is lower than in the SH, with changes of about 10% at high latitudes in the NH and 20% in
It is very difficult to detect variations for low latitude bands since the fluctuations are very small compared to the absolute value of the mean occurrence frequency. Larger variation is mainly observed for latitudes polewards of 65°. For the NH, season 2005 was particularly less active in terms of NLC occurrence. Although no clear trends are observable, the last three NH seasons (2007-2009) show a slight decrease in occurrence frequency. In the SH, years 2003/2004 and 2007/2008 saw less NLC than in other years, especially at high latitudes. It is surprising that SH season 2004/2005 has a somewhat large mean occurrence rate because, as described earlier in this chapter, the SPE event which took place in January 2005 dramatically lowered the daily occurrence rates, with lower latitudes suffering for the rest of the season from low occurrences. However, the beginning of the season saw some large NLC occurrence rates and the season itself was quite long compared to others, which can explain the high value of the mean occurrence rate.

Figure 6.17b presents the NLC radiance time series (for $\lambda = 291$ nm). The results are extremely similar to those of the occurrence rate in both hemispheres, except that the mean seasonal radiances are spread on a larger range of values. The correlation of the seasonal radiance and occurrence rate is expected and very similar to what is measured, for instance, by the SBUV instruments. The seasonally averaged NLC altitudes for both hemispheres are shown in figure 6.17c. Data for the two lowest latitude bands were omitted due to the large scatter which prevented an easy reading of the plots, without adding much information. The yearly averaged altitudes at different latitudes follow a very similar pattern in both hemispheres, also having much the same amplitude of the year-to-year variation found to lie between 0.2 and 0.5 km. The year-to-year variability is fairly well anti-correlated with the radiance and the occurrence frequency. For years of particularly large NLC activity, the altitude is lower than on average, and vice versa.

The absence of a clear trend in the NLC occurrence rate, radiance or altitude during the decreasing part of the solar cycle is noteworthy since it challenges other published works. As mentioned in DeLand et al. (2003), NLC occurrence rates and albedos gathered from multiple SBUV instruments over a period spanning 2 solar cycles showed significant anti-correlation with solar activity, larger in the NH ($R=-0.87$) than in the SH ($R=-0.65$). The SBUV occurrence frequency increased by a factor of 4-5 during low solar activity, an enormous intensification of NLC activity. It is evident that with merely seven NLC seasons available from SCIAMACHY, only a limited insight can be gained concerning the interaction between the 11-year solar cycle and NLC activity, especially compared to the 31-year record from the SBUV instruments. However, as SCIAMACHY’s period of observation coincides with an abrupt change in solar activity, an increase in NLC occurrence rates was expected to be observed. These results are however in good agreement with lidar observations of NLC at ALOMAR over the entire solar cycle 23 (Fiedler et al., 2009). The time series of lidar NLC occurrence frequency shows a significant anti-correlation with solar activity, but only for years 1997 to 2004. Afterwards, the correspondence between both variables vanishes. Analysis of NLC altitude, brightness and vertical extent time series also shows a similar behavior, which suggests that the solar signal might be masked by other sources of variability.
Figure 6.17: Time series between 2002-2009 of NLC occurrence frequency (panel A), radiance (panel B) and altitude (panel C) for both hemispheres and different latitude bands.
Table 6.1: Latitudinal variation of the standard deviation of the NLC occurrence frequency (OF), radiance and altitude within a single season. These values are representative of all years of SCIAMACHY observations.

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<th>Latitude Range</th>
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<tr>
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<td>OF [%]</td>
<td>Radiance [10^10 photons s cm^2 sr nm]</td>
</tr>
<tr>
<td></td>
<td></td>
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<tr>
<td>50°-55°</td>
<td>2</td>
<td>0.7</td>
</tr>
<tr>
<td>55°-60°</td>
<td>11</td>
<td>2</td>
</tr>
<tr>
<td>60°-65°</td>
<td>16</td>
<td>3</td>
</tr>
<tr>
<td>65°-70°</td>
<td>24</td>
<td>5</td>
</tr>
<tr>
<td>70°-75°</td>
<td>31</td>
<td>7</td>
</tr>
<tr>
<td>75°-80°</td>
<td>34</td>
<td>13</td>
</tr>
<tr>
<td>80°-85°</td>
<td>35</td>
<td>18</td>
</tr>
</tbody>
</table>

throughout the last five seasons.
In the last chapter, seven years of SCIAMACHY NLC properties have been analyzed and it was shown that the NLC occurrence frequency, radiance and altitude can provide insights concerning atmospheric processes taking place in the vicinity of the summer mesopause. This part of the thesis will focus on the NLC particle size retrieved from SCIAMACHY. A climatology of the NLC particle size covering the NH season 2002-2009 will be presented. The data will be compared to results obtained from different experimental techniques. The spatial, seasonal and year-to-year variations will be described, along with an analysis of the connection between the NLC particle radii and the latitude, altitude and local time of observation. The sensitivity of the NLC particle size on the model assumptions (PSD and shape of particle) will also be investigated. Finally, the first results of an international scientific cooperation involving SCIAMACHY will be presented, describing how consistency of the NLC particle size among various data sets can be achieved.

It was already discussed in section 5.5 that assumptions must be made concerning the PSD and shape of the NLC particles in order to obtain particle size information from SCIAMACHY’s measurements. Except when stated otherwise, the particle size retrieved assumes that the ice particle layer consists of spherical particles with a normal PSD of width $\sigma = 22$ nm. These assumptions are based on recent lidar measurements from Baumgarten et al. (2007) which determined that the width of the Gaussian PSD describing best the measurements corresponded to $\sigma = 22.2 \pm 1.6$ nm.
Figure 7.1: Mean daily NLC particle size for latitude bands 65-75° (square) and 75-83° (triangle) in the northern hemisphere summer, averaged over years 2002-2009. The particle size was calculated for a Normal PSD with $\sigma = 22$ nm. The curves were smoothed with a 5-day running mean.

### 7.1 NLC particle size climatology

Figure 7.1 presents the mean seasonal variation of NLC particle radii retrieved from SCIAMACHY measurements between 2002-2009 and for latitude ranges 65-75° and 75-83°. Both curves are smoothed with a 5-day running mean. For high latitudes, the particle size increases during the first part of the season, starting from 57 nm at -20 DTS and reaching a maximum value of $\sim 90$ nm for 35 DTS, similarly to the occurrence rate. Physically, the environmental conditions of the upper mesosphere at the core of the NLC season is expected to benefit the growth of larger particles due to the higher level of saturation and the vertical extent of the saturated region.

The NLC particle size at lower latitudes is smaller than at high latitudes, and so is the amplitude of its seasonal variation. During the first days of the season, particle size at low latitudes is similar to high latitude observations but reaches a plateau at 73 nm around solstice. After that point, the value of the particle size oscillates around that value due to natural variability and finally starts decreasing around 40 DTS to finally reach values below 60 nm at 60 DTS. The NLC particle size in the 65-75° latitude band is smaller than at high latitudes by about 10-15 nm.

The spatial and temporal variation of NLC particle size for the individual NH seasons between 2002 and 2009 is presented in figure 7.2. The contour plots show the mean NLC radii for different latitudes (binned in 5° between 60°-83°) and day of the season relative...
to solstice. The contour lines inside the figures are smoothed with a 15-day running mean to show the general behavior of the seasonal variation. Radii retrieved at latitudes lower than 60° are rare, probably because the NLC events are not strong enough to have sufficient confidence in the retrieved particle size. The particle size increases with latitude and reaches a maximum value of about 90-95 nm at 83°. With regards to temporal variability within a season, the particle size is generally smaller at the beginning and the end of the season for latitudes larger than 70°, but does not seem to reach a maximum value at a fixed time in the season. The particle size is more stable in time at higher latitudes, whereas at lower latitudes, the atmospheric conditions are not always favorable to the presence of observable NLC particles. The size of the particles, mainly found to be between 40 and 90 nm, does not change substantially from year to year. There may be an exception at high latitudes, especially during the core of the season; for instance, no particles larger than 80 nm occur in season 2009 while for season 2007, the particle size reaches almost 100 nm around day 35.

The overall mean spatial and temporal variation of the NLC particle size is presented in the lower right panel of figure 7.2. The main features are easier to see in such a representation since most of the variability specific to a given year has been filtered out, leaving mostly only seasonal variations. As for the radiance and occurrence frequency, the NLC particle radii variation throughout the season is slightly asymmetric, with a peak somewhere between 20 and 40 DTS. This shift of the peak is, however, possibly due to the large values around 35-40 DTS of the 2007 NH season.

7.2 Latitude dependence of the particle size

It is interesting to have a closer look at the population of particle sizes for different latitudes. Figure 7.3 shows the histogram distribution of particle sizes for six different latitude bands between 55° and 83°. A Gaussian function is fit through the data and the median and standard deviation of the fit are visible in each plot. As the latitude increases, a clear shift of the particle size from smaller to larger mean radius is observed. The radius changes from 64.6 nm up to 86.4 nm for a change in latitude from 55° to 83°. The width of the distribution is also largest at high latitudes, taking a value of 20.1 nm for the 80°-83° latitude interval and going down to 11.4 nm for the 60°-65° latitude band.

The variation of the particle size with latitude was already considered by von Savigny and Burrows (2007) using 2005 NH data from SCIAMACHY and Karlsson and Rapp (2006) for the SH season 2005 as measured by OSIRIS. Both studies found similar trends. Furthermore, the results from Karlsson and Rapp (2006) indicated the presence of a bi-modal structure in the histograms of the retrieved particle effective radius for high latitude observations. They interpret this second mode radius as coming from particles experiencing a second life cycle. The SCIAMACHY data set does not confirm the presence of such bi-modal structure for NH, as the particle size histograms retain a near Gaussian shape throughout the latitude ranges. Since NLC particle size cannot be retrieved by SCIAMACHY in the SH, no direct comparison
Figure 7.2: Daily averaged NLC particle size for latitudes larger than 60° in the northern hemisphere for each individual NLC season between 2002 and 2009. The seasonal variation at a given latitude was smoothed with a 5-day running mean. The contour lines inside the figures are smoothed with a 15 day running mean to show the general behavior of the seasonal variation. The mean seasonal and latitudinal variation of the particle size calculated over all years (2002-2009) is shown in the lower right panel.
7.2 Latitude Dependence of the Particle Size

Figure 7.3: Particle size histograms for various latitude bands along with a Gaussian fit through the data. Notice the evident decrease in the fit mean radius with decreasing latitude.

can be made. Results from different instruments are needed to verify whether this is a real inter-hemispheric difference, or if the presence of the bi-modal structure is an artifact of some kind. If the effect is real, it could shed light on some of the differences between the hemispheres in terms of atmospheric processes and possibly dynamics.

Figure 7.4 shows the dependence of the particle size on latitude in more detail. The small (grey) dots represent all SCIAMACHY retrieved particle sizes between 2002 and 2009. The experimental errors were omitted to improve the visibility of information on the graph. The (blue) triangles show the value of the Gaussian fit mean radius for the different latitude ranges, as were presented in figure 7.3. The (black) squares are the mean values calculated from all data points for the latitude ranges 55-60°, 60-65°, 65-70°, 70-75°, 75-80° and 80-83°. The red error bars represent the standard deviation of the radius within the latitude range while the black one is the error of the mean. All data points confirm the variation of the radius with latitude, with a larger gradient at high latitudes. For latitudes larger than 65°, the variation of the mean particle size with latitude is $1.4 \pm 0.1$ nm/deg.
Despite these results which seem unambiguous regarding the variation of the NLC particle sizes with latitude, a word of caution is required regarding the interpretation of the results. Due to the satellite’s orbit, the local time of observation changes constantly along the orbit, more abruptly for latitudes larger than 70°. Between 70° and 83°, the satellite covers 7 hours of local time. It will be shown in section 7.4 that the local time of observation has an effect on the size of the NLC particles, which makes the interpretation of the large increase of particle size with latitude equivocal.

Nonetheless, results obtained in this section are considered significant and not strongly biased due to the effect of local time, because the particle sizes sampled for this part of the study were observed on the descending part of the orbit, meaning that the time of observation varies between 10AM and 4PM local time. Measurements by Stevens et al. (2009) and Fiedler et al. (2005) show that only very little change in NLC activity (occurrence frequency, altitude and brightness) takes place between 10AM and 5PM local time. It is therefore reasonable to assume that the particle size would not be significantly changed by the local time for these observations.
7.3 Altitude dependence of the particle size

As already stated in section 6.3, according to the current understanding of the growth process of NLC particles, they form near the mesopause where saturation ratios are largest, and as they grow, they sediment under the action of gravity through the atmosphere. It is therefore reasonable to imagine that the size of the particle will be dependent on the altitude: the lower it gets, the more potential it has to grow. There is already evidence that the particle size is dependent on the altitude of observation. von Savigny et al. (2005b) used NLC particle size derived from the Optical Spectrograph and InfraRed Imager System (OSIRIS) instrument in both hemispheres and showed that, for a subset of 16 bright NLC, the particle size is dependent on the altitude of observation, with the largest radii detected at lower altitudes. Similar vertical variation of the particle size has also been measured by lidars, which have a very good vertical resolution (Baumgarten and Fiedler, 2008), and were also measured by rocket measurements (Gumbel and Witt, 1998).

The SCIAMACHY particle size data set is considerable and could be analyzed to provide further evidence of this effect and quantify the vertical variation of the particle size. Unfortunately, the coarse vertical resolution of the instrument prohibits direct correlative analysis, since limb observations are performed on a ∼ 3 km vertical grid at nearly fixed altitudes. Figure 7.5a presents the altitude and size of all particles retrieved from SCIAMACHY between 2002-2009. It can be seen that the particle observations cluster around 81 and 84 km, i.e., at the tangent heights of SCIAMACHY’s observation. The investigation of the variation of the particle size with altitude can nonetheless be carried out by dividing SCIAMACHY's observations in two categories, i.e., high and low clouds, and looking at the distribution of the retrieved particle sizes within these categories.

Figure 7.5b and figure 7.5c show the distribution of SCIAMACHY particle sizes observed at altitudes higher than 82.5 km and lower than 82.5 km, respectively. Only observations made on the descending part of the orbit are taken into account here. A Gaussian fit through the data is also shown, and the parameters of the fit are presented in each figure. The data show that high clouds, with a mean altitude of 84.04 km, have a particle size of 67.0 ± 14.3 nm, whereas NLC particles clustered near 81 km are generally larger with a mean mode radius of 83.8 ± 17.4 nm. The difference in particle size between the two ensembles is highly significant (> 99.99%) due to the sheer number of observations, and provides further confirmation that NLC particles found at lower altitudes are larger. Based on these results, the change of the particle radii with altitude is -5.3 nm/km for a normal PSD with σ = 22 nm. This is somewhat low compared with lidar measurements, which registered variations of the order of 20 nm/km at the bottom of the cloud (Baumgarten and Fiedler, 2008), and rocket experiments which observed an NLC particle size gradient of about 15 nm/km for the lowest portion of the cloud (Gumbel and Witt, 1998). On the other hand, SCIAMACHY’s results are very much in line with the ~ 5 nm/km particle size vertical gradient observed by von Savigny et al. (2005b). It should be noted however that the variation of the particle size with altitude will be strongly dependent on the PSD assumed for
Figure 7.5: Panel A: Distribution of particle sizes for NLC observations at different altitudes for all available data from SCIAMACHY. Panel B: Histogram of high cloud particle sizes, along with a Gaussian fit. Panel C: Histogram of low NLC particle radii with Gaussian fit.
7.3 Altitude Dependence of the Particle Size

Another approach to get insights into the vertical variation of the particle size is to take advantage of the daily mean particle size throughout the season and correlate it with the daily mean NLC altitude which is then not limited to values of 81 and 84 km. Figure 7.6 presents the evolution of both time series for NLC in the 75°-83° latitude band (left panel) as well as a scatter plot of the data (right panel). According to these results, the daily mean particle size is anti-correlated with the daily mean NLC altitude of observation, with a correlation coefficient of $-0.9$. There is a higher level of scatter for mean NLC altitude above $\sim 83$ km. The variation of the particle size with NLC altitude derived from this method is $-15.3 \pm 0.5$ nm/km, much larger than before. For low latitudes NLC (65°-75°), the data is not as nicely correlated (R= $-0.69$), but the estimated sensitivity of the particle radii with altitude has a value similar to what was obtained for high latitudes NLC, namely $-14.6 \pm 0.8$ nm/km. These values are much better in line with the gradients estimated by Gumbel and Witt (1998) and Baumgarten and Fiedler (2008). It is especially interesting that the values are consistent with the lidar measurements, since the PSD retrieved by Baumgarten and Fiedler (2008) is similar to the one assumed in this work. The disagreement of SCIAMACHY particle size vertical gradients estimated using two different methods is not very surprising given the limited altitude resolution. Nonetheless, it provides a range within which the particle radii variation with altitude is likely to be found.
7.4 NLC particle size for different local times of observation

The local time at which NLC are observed can have a large impact on their properties. Many different ground based instruments and models have shown that there is a strong diurnal and semi-diurnal variation of NLC occurrence and brightness (von Zahn et al., 1998; Klostermeyer, 2001; Berger and von Zahn, 2002; Fiedler et al., 2005; Chu and Gardner, 2003). More recently, satellite measurements from the Spatial Heterodyne IMager for MEsospheric Radicals (SHIMMER) instrument at low latitudes also found evidence of a variation in NLC occurrence frequencies with local time, which can change by as much as an order of magnitude (Stevens et al., 2009). The fact that there is a variation of NLC properties with local time is important because it can suggest atmospheric processes important on a diurnal and semi-diurnal timescale in the upper mesosphere. Moreover, it underlines the importance of the local time when comparing NLC properties observed by different instruments.

The fact that SCIAMACHY is in a sun-synchronous orbit limits greatly the investigation of the variation of NLC properties over the entire range of illuminated local times. The local time of SCIAMACHY’s observations, already presented in figure 4.1, shows a strong variation with latitude near the pole, and it is hence very difficult to isolate the contribution of any variation in NLC properties to the local time of observation alone. However, by using both the ascending and descending portion of the orbit and carefully selecting the latitude range, it is possible to limit the range of local times covered to one hour while observing the same latitude band. For this part of the study, two ensembles of particles are defined and the difference in their properties is attributed to the local time of observation. The latitudes in the range 50°-70° are considered, and the local times of the ascending and descending portion of the orbit are 10.30-11.30 and 20.30-21.30, respectively.

For both local times of observation, the histogram of the particle sizes ensemble is shown in figure 7.7. The NLC particle radii on the descending part of the orbit (≈ 11AM) are 6.1 nm smaller than NLC particles on the ascending part of the orbit (≈ 9PM). The difference cannot be attributed to latitude as there is less than 1° difference in the mean latitude of observation for these ensembles. The histogram of the particle size at ≈ 9PM shows a much wider distribution than for the AM observations, with a width of 26.7 nm compared with 13.3 nm for the AM particle radii. This difference is most probably the consequence of the high sensitivity of the particle size retrieval during the ascending part of the orbit due to the small scattering angles, an effect which was already discussed in section 5.5. It is also possible that the different local times of observation also affect the width of the particle size ensemble. Nevertheless, the fact that the distributions differ in terms of width does not affect the conclusion that the NLC particle size is larger near 9PM than around 11AM, a result statistically significant at the 99.9% level.

These results agree with lidar measurements from Fiedler et al. (2005) at ALOMAR. The analysis of the entire data set covering the period 1997-2003 shows that there is a variation of the NLC altitude with local time. For NLC observed at 11AM and 9PM, the altitude
7.5 Inter-seasonal variation of NLC particle size

Similarly to the NLC occurrence frequency, radiance and altitude, the particle size can also be examined for any evidence of change between 2003-2009. Figure 7.8 shows the time series of NLC particle size. The mean radius is calculated by fitting a Gaussian curve through

\[
\text{Number of detection}
\]

\[
\text{Particle size [nm]}
\]

\[
\text{Local time: 10.30 to 11.30}
\]

- \( r = 68.0 \text{ nm} \)
- \( \sigma = 13.3 \text{ nm} \)

\[
\text{Local time: 20.30 to 21.30}
\]

- \( r = 74.1 \text{ nm} \)
- \( \sigma = 26.7 \text{ nm} \)

Figure 7.7: Histogram of NLC particle size for observed between 10.30-11.30 LT (upper panel) and 20.30-21.30 LT (lower panel).

difference is in the range 0.2-0.4 km, with the 9PM clouds being lower. This is a small difference, but if one assumes that the vertical variation of the particle size estimated by SCIAMACHY is correct, this altitude difference corresponds to a particle size difference on the order of 3-6 nm. The difference in altitude is therefore able to explain the 6 nm change in particle size. The variation of NLC activity as a function of local time observed by SHIMMER in the NH is also in accordance with SCIAMACHY’s results, with a larger NLC occurrence frequency near 9PM compared to 11AM. The physical mechanism responsible for this local time variation, as argued by Fiedler et al. (2005), is most probably the consequence of atmospheric tides since the diurnal variation is recurrent through the data set spanning seven years.

7.5 Inter-seasonal variation of NLC particle size

Similarly to the NLC occurrence frequency, radiance and altitude, the particle size can also be examined for any evidence of change between 2003-2009. Figure 7.8 shows the time series of NLC particle size. The mean radius is calculated by fitting a Gaussian curve through
Figure 7.8: NLC particle size evolution between 2003-2009. The histograms encompass all particle sizes observed on the descending part of the orbit and at all latitudes.

Firstly, it can be observed that the distribution of NLC particle sizes for each year follows closely a Gaussian shaped curve. The Gaussian fit mean radius varies between 70.7 nm and 80.6 nm over the years. The mean particle size is rather constant at $r \approx 77$ nm for seasons 2003-2007 and decreases down to 70.7 nm during season 2009. The width of the Gaussian distribution does not change appreciatively over the years and is found to lie between 19.2 and 23 nm. The last three seasons exhibit all standard deviations larger than 21 nm. For all available years, the maximum observed particle size was 149 nm. Only 2% of the observations led to particle radii larger than 120 nm. Most particles (85%) are found between 40 and 100 nm.
A more detailed picture of the evolution of the particle size between 2003-2009 is presented in figure 7.9, showing the temporal behavior of the NLC particle size in five different latitude bins between 60° and 83°. The errors associated with the mean of the radii are not visible in the graph because their magnitude is smaller than 0.5 nm for all data points. The standard deviations linked to the variability of the particle size within a season and a given latitude range are left out in order to improve clarity of the figure, but are instead presented in table 7.1, along with the yearly averaged particle size within different latitude ranges for an assumed Gaussian PSD with $\sigma = 22$ nm.

Due in part to the change of the vertical scale of figure 7.9 compared with figure 7.8, a general decline of the particle size is observed since 2003. Season 2004 and 2007 show localized increases in the particle size compared to neighboring years, especially at higher latitudes, while summer season 2009 displays the smallest NLC particles retrieved up to now. Low latitude particle sizes (60-65°) display a somewhat larger year-to-year variability, although the magnitude of the variability between 2003 and 2009 for all latitude bands is similar to a large extent, with a difference of about 10 nm between the largest and the smallest seasonally averaged NLC particle size.

These results are in contradiction with earlier results derived from SCIAMACHY (Robert et al., 2009), for which an increase in particle size was observed at latitudes larger than 70° between 2003-2007 using a different retrieval technique. Although the relative changes are small in both cases (5 nm over five seasons for Robert et al. (2009), 5-10 nm for the results presented here), the fact that the direction of the trend differs in both studies indicates that the NLC particle size inter-seasonal variation is sensitive to the retrieval method.
Table 7.1: Seasonally averaged NLC particle size (in nm) along with their seasonal variation for different latitude bands between 2003-2009. The error associated with the seasonal mean is smaller than 0.5 nm.

<table>
<thead>
<tr>
<th>Years</th>
<th>Latitudes</th>
<th></th>
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<th></th>
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<tr>
<td></td>
<td>60°-65°</td>
<td>65°-70°</td>
<td>70°-75°</td>
<td>75°-80°</td>
<td>80°-83°</td>
</tr>
<tr>
<td>2003</td>
<td>68.5 ± 11.0</td>
<td>71.1 ± 15.6</td>
<td>77.7 ± 14.9</td>
<td>78.2 ± 19.0</td>
<td>85.3 ± 19.4</td>
</tr>
<tr>
<td>2004</td>
<td>75.8 ± 15.8</td>
<td>74.6 ± 15.3</td>
<td>79.9 ± 16.0</td>
<td>82.9 ± 17.7</td>
<td>88.5 ± 18.9</td>
</tr>
<tr>
<td>2005</td>
<td>65.3 ± 13.8</td>
<td>68.8 ± 14.0</td>
<td>75.9 ± 15.2</td>
<td>78.9 ± 17.5</td>
<td>88.0 ± 19.2</td>
</tr>
<tr>
<td>2006</td>
<td>67.8 ± 13.7</td>
<td>66.5 ± 16.5</td>
<td>73.6 ± 15.8</td>
<td>78.0 ± 17.6</td>
<td>85.3 ± 19.1</td>
</tr>
<tr>
<td>2007</td>
<td>62.2 ± 12.2</td>
<td>66.3 ± 18.1</td>
<td>73.9 ± 18.6</td>
<td>80.5 ± 18.9</td>
<td>88.7 ± 19.7</td>
</tr>
<tr>
<td>2008</td>
<td>61.5 ± 15.4</td>
<td>62.7 ± 19.3</td>
<td>71.7 ± 18.7</td>
<td>75.7 ± 19.6</td>
<td>82.3 ± 21.5</td>
</tr>
<tr>
<td>2009</td>
<td>59.8 ± 14.4</td>
<td>60.8 ± 19.6</td>
<td>65.9 ± 19.9</td>
<td>71.5 ± 21.7</td>
<td>78.8 ± 21.1</td>
</tr>
</tbody>
</table>

The main differences between the analysis of SCIAMACHY data for both studies involve an improved determination of the Rayleigh background for the results presented in this work, as well as the use of the M-factor database for the correction of channel 1 degradation and the calculation of the sun-normalized radiance using the daily measured solar spectrum. There is no reason to believe that the new Rayleigh background correction scheme could include a spurious change in the particle size over the years, nor that the daily solar spectrum could affect the particle size appreciatively. It was already shown that the correction to the \( \alpha \) exponent involving the change in solar irradiance over the 11-year solar cycle is on the order of 0.05 while the error on the \( \alpha \) exponent retrieval is about 3 times as large. The use of the M-factor data base could be a potential source for the change since the degradation is evolving from year to year. It was already stated before that the use of the M-factors is assumed to be an improvement over the off-line correction of the SCIAMACHY radiances which was done before, meaning that the results here should normally be more trusted. Due to the lack of certainty concerning the source of the change in these results compared to previous results, a cautious approach is favored and it will simply be stated that more work is needed to determine whether this decrease in NLC particle size is real or an artifact of the method. Based on the occurrence frequency time series though, which is largely unaffected by a change in channel 1 degradation, the last three NLC seasons have shown a decrease in NLC activity which could explain the results from the particle size analysis. It should be noted that the seasonal evolution of the particle size for each year is unaffected by this small change, so that previous results are still valid.
7.6 Effect of shape and PSD assumptions on the retrieved particle size

It has been repeatedly stated in this work that, in order to retrieve NLC particle size from SCIAMACHY’s spectral measurements, assumptions must be made on the particle size distribution within the cloud and the shape of the particles. All the results presented up to now assumed that an NLC is reasonably modeled by an ensemble of spherical ice particles with a normal PSD and a width $\sigma = 22$ nm, based on evidence from lidar measurements. However, one might wonder what the consequences of using a different PSD or assuming a different particle shape would be on the magnitude of the particle size. In this section, the SCIAMACHY data set is reprocessed by changing these assumptions and the resulting change of the particle size is presented.

Firstly, the particle shape is kept constant (sphere) while the PSD is changed, so as to see the effect of changing only one parameter at a time. The evolution of the mean particle size over the NH summer season is therefore computed based on Gaussian PSD with $\sigma = 14, 22, 30$ nm as well as a lognormal PSD with $\sigma = 1.4$. The results of this reprocessing, shown in figure 7.10a, indicate that regardless of the width of the Gaussian PSD, the temporal evolution is similar throughout the season which implies that the effect of changing $\sigma$ on the particle size is systematic. The magnitude of the change can be very large, depending on which PSD are compared. The difference between particle size is also greater for larger values of $\sigma$: the difference in size for $\sigma = 22$ nm and $\sigma = 30$ nm is about 20 nm whereas it is only 12 nm for $\sigma = 14$ nm and $\sigma = 22$ nm. The lognormal distribution shows slightly
less variation throughout the season compared to the Gaussian PSD, but is nonetheless in rather good qualitative agreement with a Gaussian PSD of 30 nm width. It should be noted that the amplitude of the change throughout the season differs depending on the $\sigma$ value. For the normal PSD, the difference between the lowest and largest particle size throughout the season is 30, 42 and 45 nm for $\sigma = 14$, 22 and 30 nm respectively. All the results of this paper were based on the assumption that NLC can be modeled with a constant PSD, regardless of the time of the season, height and latitude. It is clear from the analysis presented in figure 7.10a that, if this assumption is incorrect, the particle size retrieved and the temporal and spatial trends will vary greatly from what was presented, a problem also pointed out by Rusch et al. (2008).

It was already mentioned in section 3.3 that there is evidence of non-spherical NLC particles. In order to investigate the effect of this parameter on the retrieved particle size, a reprocessing of the data was done assuming spheroid-shaped particles with axial ratios 0.2, 1.0, 5.0 and cylinder-shaped particles with an axial ratio of 1.0. The sensitivity of the retrieved size to the assumed particle shape is presented in figure 7.10b. The particle size corresponds to the volume equivalent sphere radius for randomly oriented particles. These results show that particle sizes obtained assuming oblate particles are smaller than for prolate particles, a result which goes in the same direction as Baumgarten et al. (2007). It is also interesting to see that irrespective of the shape of the particle (spheroid or cylinder), the results for an axial ratio $AR = 1$ are exactly the same. Similarly to the effect of the PSD width, the difference in particle size between the various assumed particle shapes is generally systematic, with only few changes in the difference of the time series during the season. The change of $\sim 5 – 10$ nm in particle size from oblate ($AR = 0.2$) to sphere is smaller than the difference between prolate ($AR = 5$) and spheres, which amounts to $\sim 15 – 20$ nm. The difference between the minimum and maximum daily average particle size changes according to the assumed shape. This difference amounts to 60, 47 and 48 nm for the prolate, sphere and oblate spheroids respectively. The magnitude of the seasonal variability is therefore affected by the shape of the assumed particle, but temporal trends should not be significantly affected by the assumed shape of the particle, given that the shape does not change appreciably throughout the season.

### 7.7 Comparison of SCIAMACHY’s particle size with independent data sets

Besides the temporal variation of the particle size, a topic which has not yet been addressed is the comparison of the average particle size with other works. Table 7.2 presents some results concerning NLC particle size published in the course of the last decade, along with the assumptions made for the retrieval, the observation method used and the location of observations. The rightmost column contains the mean particle size as retrieved by SCIAMACHY using the corresponding assumptions and geographical extent, for all years of
Table 7.2: Comparison of SCIAMACHY’s retrieved NLC radii with some particle sizes published during the last decade. The table also shows the assumptions made for the retrieval, the experimental method used and the location at which measurements took place. The rightmost column presents SCIAMACHY’s seasonally averaged results for the corresponding latitudes, calculated using the same assumptions as the particle size compared with.

<table>
<thead>
<tr>
<th>Reference</th>
<th>Method</th>
<th>Assumptions</th>
<th>Location</th>
<th>Radius (nm)</th>
<th>R_{SCIA} (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lumpe et al. (2008)</td>
<td>POAM</td>
<td>6th moment of lognormal PSD</td>
<td>60 – 65°S</td>
<td>52 ± 17</td>
<td>51.8 ± 11.0 (NH)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Converted to Lognormal, σ = 1.4</td>
<td>60 – 65°S</td>
<td>35.5 ± 12</td>
<td></td>
</tr>
<tr>
<td>Rusch et al. (2008)</td>
<td>SNOE</td>
<td>Normal, σ = 14 nm</td>
<td>62.5°N-82.5°N</td>
<td>15-45</td>
<td>84.6 ± 8.5 (NH)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>72.5°S-77.5°S</td>
<td>5-25</td>
<td></td>
</tr>
<tr>
<td></td>
<td>SME</td>
<td></td>
<td>72.5°N-77.5°N</td>
<td>20-65</td>
<td>84.2 ± 6.2 (NH)</td>
</tr>
<tr>
<td>Baumgarten et al. (2007)</td>
<td>LIDAR</td>
<td>Gaussian, σ = 22.2 nm</td>
<td>69°N</td>
<td>41 ± 4</td>
<td>71.3 ± 20.0</td>
</tr>
<tr>
<td>Karlsson and Rapp (2006)</td>
<td>OSIRIS</td>
<td>Effective radii</td>
<td>70°S-90°S</td>
<td>65-75</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>Converted to Lognormal, σ = 1.4</td>
<td></td>
<td>35-45</td>
<td>54.0 – 60 ± 12 (NH)</td>
</tr>
<tr>
<td>Alpers et al. (2000)</td>
<td>LIDAR</td>
<td>Lognormal, σ = 1.5 – 1.6</td>
<td>54°N</td>
<td>20-27</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>Converted to Lognormal, σ = 1.4</td>
<td>54°N</td>
<td>≈ 35</td>
<td>52.5 ± 16.0</td>
</tr>
<tr>
<td>von Cossart et al. (1999)</td>
<td>LIDAR</td>
<td>Lognormal, σ = 1.42</td>
<td>69°N</td>
<td>51 ± 21</td>
<td>53.0 ± 12.6</td>
</tr>
</tbody>
</table>

Overall, SCIAMACHY particle radii retrieval seems to overestimate the radii compared to the results of other studies, which is especially clear for the investigation done by Rusch et al. (2008) using SNOE and SME measurements. Except for this result, the discrepancy between SCIAMACHY’s retrieved particle size and results from other instruments is between 10-20 nm, which is quite large assuming NLC mean particle size below 100 nm. Some of the results cannot be directly compared though since they were not measured during the same season or hemisphere (Lumpe et al., 2008; Karlsson and Rapp, 2006). However, it is unlikely that seasonal variability and inter-hemispheric differences could explain such a large deviation. Comparison of satellite retrieved radii and lidar measurements must also be done with care, due to the very different atmospheric volume sampled by both instruments. A recent study (von Savigny et al., 2009) shows that, provided the differences in spatial as well as vertical resolution between SCIAMACHY and lidars are taken into account, very good agreement is found when comparing particle sizes.

It is generally difficult to obtain agreement of particle size data sets retrieved using different experimental techniques, especially since the retrieval usually involves different methods, wavelengths and assumptions. In order to bring together information on the NLC particle size from different data sets and establish the state of the art in the measurement of ice particle sizes, a recent working group involving (currently) five different measuring platforms has been established. Table 7.3 describes briefly the different instruments taking part in the NLC particle size working group, comprising mostly satellite missions (CIPS, OSIRIS, SOFIE, SCIAMACHY) and a ground-based lidar operated in ALOMAR.
A first workshop took place with the objective of determining whether there is a set of particle size distribution assumptions which can bring the various data sets in agreement. The approach chosen for the comparison involved first selecting a subset of NLC events which could be detected by all instruments and limiting the range of latitudes covered to \(69^\circ \pm 2.5^\circ\), where the lidar instrument performs its measurements. Then, in an iterative session, various assumptions concerning the shape and PSD of the ice particles ensemble were decided upon and the particle sizes retrieved with these assumptions were compared.

**Table 7.3:** Description of the different instruments taking part in the NLC particle size working group.

<table>
<thead>
<tr>
<th>Instrument</th>
<th>Wavelength [nm]</th>
<th>Scattering angles [km³] (at 69°)</th>
<th>Retrieval technique</th>
<th>Sampled volume [ km³ ]</th>
<th>Local time</th>
</tr>
</thead>
<tbody>
<tr>
<td>SOFIE</td>
<td>330-5000</td>
<td>0</td>
<td>Color ratio</td>
<td>2250</td>
<td>24</td>
</tr>
<tr>
<td>ALOMAR LIDAR</td>
<td>355, 532, 1064</td>
<td></td>
<td>Color ratio</td>
<td>0.4</td>
<td>all</td>
</tr>
<tr>
<td>CIPS</td>
<td>265</td>
<td>30-150</td>
<td>Phase function</td>
<td>338</td>
<td>13.5, 22.5</td>
</tr>
<tr>
<td>OSIRIS</td>
<td>275-310</td>
<td>70-100</td>
<td>Spectrum</td>
<td>(18 \times 10^3)</td>
<td>8, 17</td>
</tr>
<tr>
<td>SCIAMACHY</td>
<td>264-300</td>
<td>22-60</td>
<td>Spectrum</td>
<td>(10^6)</td>
<td>11.5, 20.5</td>
</tr>
</tbody>
</table>

Figure 7.11: Panel A: Particle size histograms and seasonal variation obtained from the different instruments during the particle size workshop when assuming that ice particles are spheroids with \(AR=2\) and have a normal PSD with \(\sigma=14\) nm. Panel B: Same as A but assuming that ice particles are spheroids with \(AR=2\) and have a PSD for which \(\sigma = r/2\) nm. Figure courtesy of Scott Bailey.
Figure 7.12: Variation of the retrieved width of the PSD as a function of the retrieved NLC particle size for different portions of a NLC, from lidar measurements. Figure courtesy of Gerd Baumgarten.

Figure 7.11 shows the first comparison done during the workshop clearly presenting the divergence of the different data sets if the NLC particle sizes were retrieved assuming spheroid particles with AR = 2 and having normal PSD with $\sigma = 14$ nm. By the end of the workshop however, after much discussion and several attempts to find assumptions which could explain the different measurements, a satisfactory level of agreement was reached. Figure 7.11b shows the results obtained by assuming spheroids ice particles with AR = 0.5 and having a special PSD for which the width of the distribution changes with the radius ($\sigma = r/2$). Particle sizes from different data sets become consistent when retrieved with these assumptions, as can be seen from the histogram of figure 7.11b, and the temporal variation of the various data sets is also consistent.

Besides the good agreement of the various data sets, there is strong evidence from lidar measurements that icy particles do possess a distribution for which $\sigma = r/2$ (Baumgarten et al., 2009). As lidar measurements make it possible to retrieve both the particle size and the width of the PSD, this information can be used to understand the connection between both parameters. Figure 7.12 shows the probability distribution of the PSD width as a function of the particle size retrieved based on lidar measurements for different portions of the cloud relative to the beak brightness. It is clearly observed that there is a tendency for the PSD in all regions of the cloud to assume a width which is roughly half the radius. The value of the ratio is actually closer to $\sigma = r/2.5$, but as a first approximation $\sigma = r/2$ seems to be effective in bringing all data sets together.

Since these assumptions seem to bring consensus to the interpretation of all NLC measurements, it is interesting to see how the SCIAMACHY data set presented in this chapter changes accordingly. Figure 7.13 shows the climatology of NLC particle size calculated between 2002-2009 with the assumptions of spherical particles and normal PSD with $\sigma = 22$ nm (left panel) and spheroids with AR=0.5 and PSD with $\sigma = r/2$ (right panel). There are clear differences in the magnitude of the particle sizes derived using both assumptions, as was expected from the sensitivity studies presented in section 7.6. The new assumptions produce smaller particle sizes, with a maximum particle size of about 60 nm found at the highest latitudes. The pattern of the seasonal variation, however, is very similar to what was obtained before. The increase in particle size from $\sim 60^\circ$ to $83^\circ$ has a magnitude of 20 nm, which is about half as much as what was observed with the previous assumptions.
Figure 7.13: Comparison of the SCIAMACHY particle size climatology obtained assuming spherical particles and a normal PSD with $\sigma = 22$ nm (left panel) and spheroids with axial ratio 0.5 and a PSD where the width obeys the relation $\sigma = r/2$. 
If you want the number 216 in the world you will be able to find it everywhere. 216 steps from your street corner to your front door, 216 seconds you spend riding on the elevator. When your mind becomes obsessed with anything, you’ll filter everything else out and find that thing everywhere. 320, 450, 22...whatever. You’ve chosen 216 and you’ll find it everywhere in nature. But, Max, as soon as you discard scientific rigour, you are no longer a mathematician, you’re a numerologist.

Sol Robeson, Pi (1998)

27-day solar signature in NLC properties

It seems self-evident to claim that solar activity has an effect on the state of the atmosphere. After all, it is its main source of energy, at the root of its dynamics and chemistry. But the existence of a link between the 27-day periodicity in solar activity and meteorological parameters is not all that obvious because the fluctuations of the solar energy output on that timescale are relatively minor and their potential impacts on the earth’s atmosphere may be easily masked by the short-term variability of the weather system (Tsiropoula, 2003).

Recent studies on the subject however contain evidence that the 27-day solar variability does affect the state of the atmosphere, from the troposphere to the thermosphere. It has been known since quite a while that there is a 27-day signal in the stratosphere, especially in O₃ (Hood, 1986; Fleming et al., 1995; Dikty et al., 2009) and temperature (Hood, 1986). The thermospheric density can be considerably modulated during a 27-day cycle, by up to 20% of its mean value (Rhoden et al., 2000). This is not surprising as the region of the earth’s atmosphere above 200 km is considered to be the most sensitive single gauge of solar activity (Jacchia, 1964). Ionospheric parameters such as total electron content (TEC) are also changed by the sun rotational cycle (Oinats et al., 2008). There are few studies on the presence of a 27-day signal in the mesosphere tough. A very recent paper using 18 years of mesospheric OH temperature at 87 km showed that there is such a signal and that it is strongly modulated by the 11-year cycle (Höppner and Bittner, 2009). A review of the mesospheric temperature response to the solar rotational variability is also given by Beig et al. (2008), in which some evidence of a 27-day variation of mesospheric temperatures is highlighted. Finally, there are even indications of a tropospheric response to the 27-day signal in the form of a modulated tropospheric cloud cover in the Western Pacific warm pool region (Takahashi et al., 2009).
These studies are of considerable importance for the climate research community, because the effect of solar variability on the climate forcing over longer (>22 years) timescales is possibly of significance for climate models. Based on these observations, mechanisms for the interaction between solar activity modulation and atmospheric parameters can be verified and the impact of long-term change of the solar output can be estimated by global climate models (GCM). There have been but few measurements of atmospheric variables in the mesosphere compared to the troposphere and lower stratosphere. This is partly due to the difficulty of performing measurements in this region, which is directly accessible only to rockets. Although the situation has considerably improved in the recent decades, with space-borne missions dedicated to the study of the earth's middle and upper atmosphere and the advances made in ground-based technology, information on the mesosphere and the summer mesopause is scarce. This is especially true of data sets spanning several 11-year solar cycles.

An indirect indicator of the state of the summer mesopause are NLC properties. An advantage of using these compared to other atmospheric variables is the availability of long-time series of their properties, with more than 40 years of ground-based observations (Romejko and Dalin, 2003) and 30 years of satellite measurements (Shettle et al., 2009). Since NLC are very sensitive to changes in mesospheric temperature, and water vapor to a lesser extent, changes in these attributes are expected to be mirrored in the modulation of NLC properties. Therefore, if a 27-day solar signal is present near the summer mesopause, either in temperature or water vapor, it should be apparent in NLC properties.

It is already known that NLC are impacted by the 11-year solar cycle (Gadsden, 1985; Thomas et al., 1991; DeLand et al., 2003; Hervig and Siskind, 2006), but the mechanisms responsible for this variation have not been thoroughly substantiated. A legitimate question would be whether there is also a response of NLC to the 27-day solar variation. This is an interesting topic because it can help to understand what kind of role the solar variability can play in the summer mesosphere region. If present, it would be intriguing to see if the 27-day variation scales with the 11-year modulation of NLC properties, suggesting similar mechanisms behind both variations. The time of response of NLC properties on the 27-day solar cycle could also further restrain the mechanisms which might potentially explain the sun-NLC interaction.

In this chapter, the effect of the 27-day solar flux variation on NLC occurrence frequency is investigated using NLC data sets from the SCIAMACHY and SBUV instruments. Cross-correlation plots of solar Lyman-α irradiance and NLC occurrence frequency anomalies are presented for years 2002-2009 for SCIAMACHY and 1979-2006 for SBUV. Results obtained through a superposed epoch analysis of the solar forcing on NLC occurrence frequency are also presented in order to substantiate the relationship between these geophysical parameters. In connection with this analysis, an investigation of the response of MLS mesospheric temperatures and H₂O mixing ratios to the 27-day cycle is carried out for the summer seasons 2005-2008 in both hemispheres.
8.1 Methodology and data analysis

The challenge of this study lies in the detection of the 27-day solar signal during the short NLC season. If only the season’s core containing approximately 90% of the total NLC detections is examined, about 70 days are left to detect a 27-day signal. Moreover, as described in section 3.3, many other processes with no direct link to the solar irradiance impact on NLC, and so the search for a connection between NLC and solar activity will invariably be affected by these. The proxy for NLC activity must therefore be chosen with care so as to reflect best the effect that the solar radiation could have on NLC and maximize the population sampled. Among the many alternatives of possible NLC properties, the daily occurrence frequency, averaged zonally and over a latitude range of 60-80° was chosen as a good indicator of NLC activity. It has the advantage of being simple, easily retrieved with few assumptions made and available from both SCIAMACHY and SBUV. Another proxy which will be analyzed in this work is the daily mean NLC albedo, used in many SBUV studies (DeLand et al., 2007). It is strongly correlated with the occurrence frequency for SBUV data, but its dynamic range is larger than for the occurrence rate, which is limited to the [0-1] range. For SCIAMACHY, the daily averaged NLC radiances at 291 nm will be used as a proxy with larger dynamic range.

Anomalies

The NLC properties vary strongly during the 12 weeks of a typical NLC season. The removal of this seasonal variation is imperative for the appropriate computation of the anomalies which will be used for the correlation analysis. Different approaches exist to remove the seasonal component in a time series, and it is important to make sure that the results obtained are not dependent on that particular pre-processing of the data set.

One popular method to calculate anomalies is to transform the original signal in the frequency domain, multiply the spectra by an appropriate high-pass filter and transform that signal back to the time domain. Another possibility is to calculate the entire seasonal running mean of the signal, using a given boxcar length, and remove it from the original signal. The length of the boxcar is chosen so as to get rid of the large seasonal modulation while the variations on shorter timescales are ideally not altered. A comparison of anomalies calculated using different boxcar lengths shows that the results are only slightly dependent on the procedure employed. Figure 8.1 presents examples of anomalies calculated using 27, 35 and 50-day moving averages for SBUV and SCIAMACHY NLC occurrence frequencies. It is apparent that SCIAMACHY anomalies are more sensitive to the boxcar length than the SBUV data. This is a result of the larger sensitivity of SCIAMACHY to NLC, which in turn leads to a larger dynamical range of the NLC occurrence frequency and thereby an enhanced seasonal component which can be more difficult to remove. However, for both instruments, the amplitude of the anomalies only change by a few percent and the overall
Figure 8.1: Anomalies calculated using the moving average method with a 27, 35 and 50-day boxcar for a) SBUV (NH 1981) and b) SCIAMACHY (NH 2004) data. While the amplitude of the anomalies can change by a few percent, this will not significantly affect the correlation analysis since the spectral features remain the same across all curves.

The running mean approach is favored in this work because it handles missing data points without difficulty and is therefore straightforward to apply to all data sets. Anomalies are calculated using a 35-day boxcar as it was also employed in a similar study by Hood (1986) to analyze the response of ozone to short-term changes of solar UV flux. It should be noted that in order to smooth the anomalies, a final 5-day moving average is applied after the removal of the seasonal signal, which can introduce a small shift of the signal in the time domain. Cross-correlation analysis of anomalies produced using different smoothing parameters showed that this sometimes leads to an apparent phase lag of ±1 day.

Statistical significance of correlation

Since this part of the work deals with the investigation of a connection between NLC occurrence and solar activity as well as other physical variables such as temperature and water vapor, cross-correlation between different time series is a tool which will be used extensively. The statistical significance of the correlation cannot, in this case, be computed using Student’s t-distribution because adjacent observations of the time series are usually not independent from each other. Different techniques exist to evaluate the significance of the correlation between data sets (e.g., Burnaby, 1953; Zwiers, 1990), but few are able to deal appropriately with serial correlation.
Ebisuzaki (1997) developed a simple method for the accurate computation of the correlation coefficient’s statistical significance when dealing with autocorrelated time series. The procedure is similar to bootstrapping (Bühlmann, 2002) insofar as it takes the original data set and produces an ensemble of synthetic time series with the same mean and variance as that of the original data set by random resampling. This ensemble of random time series can then be used to produce a statistical distribution of the correlation coefficient between the original time series and another signal of some kind, the Lyman-α irradiance in our case. This finally provides an estimate of the likelihood of obtaining the correlation coefficient calculated for the original time series. However, mere permutation of the data set in the time domain does not account for its original serial correlation. Ebisuzaki realized that and converted the original data set to the frequency domain, produced each sample of the synthetic ensemble by adding a random phase to each frequency component and converting back to the time domain. The data set obtained in this fashion has the same autocorrelation as the original one, which is what is needed for an unbiased statistical test for time series.

For the seasonal comparison of NLC anomalies and Lyman-α time series, the confidence level of the statistical test for the correlation coefficient is set to 90%. While this figure may seem low in comparison with similar research, its use is justified by the fact that not all NLC occurrence anomalies are expected to be explained by the Lyman-α variation alone. Many different mechanisms can be responsible for variation of the NLC occurrence throughout the season, but a correlation of some kind should be present. Therefore, a 90% confidence level seems fair in this context.

Superposed epoch method

The response of the NLC properties to the 27-day cycle is not expected to be larger than other competing processes. Hence the situation is such that the response signal is potentially masked by random noise. The superposed epoch analysis, also sometimes referred to as compositing or Chree analysis, is a method used to investigate the response of a geophysical parameter in relation to some forcing, and is particularly well-suited for this kind of situation. Since its introduction by Chree (1912, 1913), this type of analysis has been used extensively and in many different disciplines (Severny et al., 1976; Scherrer et al., 1979; Mass and Portman, 1989; Luhr et al., 1998), especially in connection with solar periodicities.

Figure 8.2 visually describes the procedure which consists in defining so-called epochs in the response time series based on key events taking place in the forcing time series. The purpose of this study is to see whether the 27-day solar irradiance variation has an effect on the NLC occurrence frequency. All key events in the Lyman-α anomaly time series are therefore catalogued (i.e., whenever a clear maximum or minimum in the amplitude of the 27-day cycle is present) and the corresponding response in NLC occurrence frequency is studied using a window of data points centered on each key date. A window containing 61 days is used so that one can observe the NLC response 30 days before and 30 days
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Figure 8.2: The method of superposed epoch analysis. Panel A: Situation where a signal is acted upon by a forcing investigated (black arrows) while other random forcing (grey arrows) can also impact on the signal under study. Panel B: Definition of key events of the forcing as well as windows around these key events. Panel C: Averaging the response of the signal with respect to the key events. One can obtain the response of the signal as a function of time in the vicinity of the key event.

after the key event. A matrix is constructed using this information by entering the NLC response as a function of time relative to the key date in each row (the central column corresponds to the key date, i.e. day 0). For every key event defined, a new row is added. In order to remove from the NLC occurrence anomalies the random noise contributed by other processes such as wave activity or possible interhemispheric coupling, all of these events are averaged column-wise, i.e. for each day relative to the key date. Given a large enough sample of key events, the causal response of the NLC occurrence frequency to the solar forcing, if indeed present, should emerge in the average while random noise in the data should decrease. The window will allow us to see what is the mean phase lag of the response relative to the forcing. In order to test if the calculated average response is statistically significant, a bootstrapping approach is used, where a large number of key date samples are randomly generated and a probability distribution of the average responses of the NLC occurrence frequency anomalies are calculated in this way.

8.2 SCIAMACHY time series

The NLC occurrence rate anomalies measured by SCIAMACHY between 2002-2009 for both hemispheres are presented in Figure 8.3 along with the Lyman-α anomalies (dashed line). Cross-correlation plots of both time series are also shown for each year, with the horizontal dashed lines in the cross-correlation plot designating the correlation coefficient statistically significant at the 90% confidence level (both positive and negative correlation).

For most years in the northern hemisphere, a strong anti-correlation between Lyman-α and SCIAMACHY NLC anomalies is observed, particularly marked in 2004 and 2005. Statistically significant anti-correlation is found for years 2004, 2005 and 2007 (for 0 day lag), which indicates that there is less than 10% probability that this is simply coincidental. The results for the southern hemisphere (SH) show that seasons 2002-2003, 2004-2005,
8.2 SCIAMACHY TIME SERIES

Figure 8.3: Time series of SCIAMACHY NLC occurrence anomalies (solid) and Lyman-α irradiance anomalies (dashed) for different years and the associated cross-correlation function for the northern hemisphere (panel A) and the southern hemisphere (panel B). Note that the vertical scale changes from year to year. The dashed lines in the cross-correlation plots represent the positive and negative correlation coefficient significant at the 90% confidence level, calculated using the method explained in section 8.1.

2008-2009 have a statistically significant anti-correlation between solar activity and NLC occurrence, although seasons displaying a positive correlation between Lyman-α and NLC anomalies are also present (2003-2004 and 2006-2007).

The minimum correlation coefficients are found to be at different phase lag depending on the season, sometimes happening after the peak in solar activity (positive lag) and sometimes occurring even before (negative lag). While a negative lag seems to contradict a causal relationship between the NLC anomalies and solar activity, other processes might take place which could alter the cross-correlation analysis and especially the location of the phase lag minimum. Overall, the cross-correlation minimum is in the vicinity of 0 day, which suggests that short-term solar variability affects NLC on a very short timescale.

The amplitude of the anomalies relative to the mean NLC occurrence frequency during the core of the season is 5-10% in the NH and 15-25% in the SH. It is observed for both hemispheres that, while the NLC occurrence rate anomalies are found to vary mostly between -0.1 and 0.1, the amplitude of the Lyman-α anomaly changes from year to year. It seems therefore that either the sensitivity of NLC occurrence rates to solar activity also changes from year to year or that the signal response amplitude caused by the solar activity does scale with the proxy, but variability due to other processes does not. Nonetheless, the
effect of solar activity on NLC occurrence is expected to appear more conspicuously for seasons when the variation of the Lyman-α anomaly is large. Figure 8.4 shows the minimum correlation coefficient (within a ± 1 day phase lag) against the standard deviation of the Lyman-α anomalies throughout the season for both hemispheres. The NH data show that for years with large seasonal variations of the Lyman-α anomaly, a stronger anti-correlation is found between both time series. This effect is depicted more clearly by the dashed line, which is simply a linear fit through the NH data. A similar analysis for the SH data set shows a similar albeit weaker relationship, except for season 2003-2004 during which the Lyman-α irradiance varied strongly.

8.3 SBUV time series

In order to substantiate the connection between NLC occurrence and solar activity established with the SCIAMACHY time series, the same analysis as in section 8.2 is carried out for 28 years of SBUV data. As both time series overlap, it is also possible to see if the SCIAMACHY measurements are reproduced in the SBUV analysis. This will provide a sound basis for rejecting the possibility that the correlation is due to an instrumental artifact.

Figure 8.5 presents the NLC occurrence frequency anomalies (solid line) alongside the Lyman-α anomalies (dashed line) and a cross-correlation plot of both time series, for years 1979-2006 in the NH. It is important to bear in mind the year-to-year variability of the vertical axis range of the anomalies, which maximizes the visible amplitude of both time series. Similarly to SCIAMACHY results, many years show statistically significant
Figure 8.5: Time series plots of northern hemispheric SBUV NLC occurrence anomalies (solid) and Lyman-α irradiance anomalies (dashed) for different years and the associated cross-correlation function.
anti-correlation, and the phase lag of the minimum correlation can be positive or negative and averages out to -0.3 days for all statistically significant negative correlations. NH seasons 1980, 1986, 1990, 1992, 1995, 1996, 1998, 2000, 2003, 2005 and 2006 show particularly clear anti-correlation between NLC anomalies and solar activity. Note also that for years of overlapping measurements between SCIAMACHY and SBUV, though there are slight discrepancies between the amplitude of both instrument’s anomalies, the correlation analysis leads to similar results with significant anti-correlation for years 2003 to 2005. There are also years with positive correlation between both anomalies, the most noticeable being 1984, 1988, 1994 and 1997. The standard deviation of the anomalies corresponds to about 20% of the NLC occurrence rate in the NH.


Figure 8.7 shows a scatterplot of the minimum correlation coefficient within a ± 1 day phase lag interval as a function of the Lyman-α irradiance anomaly standard deviation for SBUV data in both hemispheres. Data points enclosed in a square symbol designate years for which the anti-correlation is statistically significant. One interesting feature of these plots is that the vast majority of data points has negative correlation coefficients, with mean correlation of -0.31 and -0.24 for the NH and SH respectively. However, unlike SCIAMACHY results in Figure 8.4, no clear linear relationship between the magnitude of the correlation and the amplitude of the Lyman-α anomalies seasonal variation is observed.

### 8.4 Superposed epoch analysis

Apart from the seasonally centric approach used previously in the cross-correlation analysis, the data sets are also analyzed using the superposed epoch method described in section 8.1. This procedure takes advantage of long time series to improve the signal to noise ratio of the NLC occurrence frequency anomaly response to solar forcing.

The analysis is carried out using two different types of events to determine the key dates in the time series: maximum and minimum of the Lyman-α anomalies. Only extrema with a clear localized peak were chosen in order to obtain unambiguous responses. Sixteen such events take place throughout the SCIAMACHY data set in the NH, and eighteen in the SH. The mean response of the SCIAMACHY NLC occurrence frequency anomaly to solar forcing is shown in Figure 8.8 for the NH (panel B) and the SH (panel D). The solid (dashed) curve
Figure 8.6: Time series plots of southern hemispheric SBUV NLC occurrence anomalies (solid) and Lyman-α irradiance anomalies (dashed) for different years and the associated cross-correlation function. Note that season 1982-1983 is missing.
Figure 8.7: Minimum of the cross-correlation function of SBUV NLC occurrence with Lyman-α anomalies in the phase lag interval [-1, 1] as a function of the Lyman-α anomaly standard deviation during the season for the NH (left) and the SH (right). Data points outlined by square symbols represent anti-correlation statistically significant at the 90% confidence level.

represents the response to conditions of maximum (minimum) solar activity. The figure also presents the average Lyman-α forcing in both hemispheres (panels A and C). The response of northern hemispheric NLC occurrence frequency to maximum Lyman-α condition is at a local minimum for 0-day phase lag. This is in good agreement with the anti-correlation of solar forcing and NLC occurrence frequency previously presented in sections 8.2 and 8.3. This result is statistically significant at the 97% confidence level. Figure 8.8 also displays the result of a spectral analysis of the different signals, obtained with the method of superposed epoch, using a fast-Fourier transform (FFT). It is interesting to note that all signals exhibit a clear 27-day period, accompanied by a 13.5-day period in some cases. This is another confirmation of a solar signature in NLC occurrence frequencies. Not very surprisingly, the response of NLC to a minimum in solar activity is consistent with this picture, with a phase lag of 0 day, a recurrence period between 25-30 days and a statistically significant anti-correlation at the 96% confidence level. Results in the SH show some characteristics of the NH curves such as a 27-day signal and a general anti-correlation with the Lyman-α irradiance, but are noisier. For conditions of maximum solar irradiance, a local minimum in NLC occurrence frequency anomalies is obtained at a phase lag of +4 days. The situation is hazy around the occurrence of the solar minimum, with a local minimum at 0-day lag and local maxima at ±5-day lag. There is a strong negative response at phase lags of about ±13 days which is related to the maximum in the solar activity and is statistically significant at the 98% confidence level.

The results of the analysis for the longer SBUV time series, containing 66 epochs in the NH and 62 in the SH, are presented in figure 8.9. NLC response in the NH provides additional support to the hypothesis that there is an anti-correlation between the 27-day solar forcing and NLC occurrence. The negative-correlation is statistically significant at the 99.9% confidence level for the case of maximum solar activity and 99.2% for minima in the Lyman-α anomalies. The improved statistics of the SBUV data show that similar results
Figure 8.8: Averaged Lyman-α forcing over all selected epochs in the NH (panel A) and SH (panel C) along with the corresponding SCIAMACHY NLC occurrence frequency anomaly response using the superposed epoch analysis in the NH (panel B) and SH (panel D). The solid (dashed) line shows the response to a maximum (minimum) irradiance of the 27-day solar forcing for the 60°-80° latitude range. Also shown on the right hand side is the fast-Fourier transform (FFT) performed on the signal from the left. The 27-day signal is dominant in all figures and the 13.5 day signal can also be observed in some cases.
Table 8.1: Level of significance of the NLC response to the epochs of maximum and minimum in the 27-day Lyman-α variation for both hemispheres and different data sets.

<table>
<thead>
<tr>
<th></th>
<th>Northern Hemisphere</th>
<th>Southern Hemisphere</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Min</td>
<td>Max</td>
</tr>
<tr>
<td>SCIAMACHY occurrence</td>
<td>97%</td>
<td>96%</td>
</tr>
<tr>
<td>SBUV occurrence</td>
<td>99.9%</td>
<td>99.2%</td>
</tr>
<tr>
<td>SBUV albedo</td>
<td>99.6%</td>
<td>97.3%</td>
</tr>
</tbody>
</table>

are obtained in the SH, although the phase-lag of the peak response is not easily identified around the key event and spans a period of ±1 day in the case of solar maximum event, and ±3 days for solar minima. The anti-correlation is statistically significant at the 99.3% confidence level for maximum solar forcing and 98.7% for minimum. All signals also exhibit a strong 27-day period in addition to a weaker 13.5 day feature.

It was already stated in the introduction of this chapter that the NLC albedo measured by SBUV instruments is strongly correlated with the frequency of occurrence. A superposed epoch analysis on the SBUV NLC albedo time series has also been conducted and the results are, as expected, extremely similar to the analysis using the SBUV NLC frequency of occurrence. A clear anti-correlation was found for both hemispheres and for both the maximum and minimum in Lyman-α. The statistical significance of the anti-correlation found using the superposed epoch analysis for the SBUV time series (albedo and occurrence frequency) and SCIAMACHY (occurrence frequency) in both hemispheres is presented in table 8.1. All the results from the analysis show that the anti-correlation between the 27-day solar cycle and NLC properties are statistically significant at least to the 95% confidence level, except for the response of SCIAMACHY NLC occurrence frequency anomalies in the SH. The shorter time series from SCIAMACHY is probably to blame for this, with less than a third key of events present in the time series compared to SBUV. In any case, using data sets spanning different periods, different hemispheres and coming from different instruments, the evidence for the presence of a 27-day signal in NLC properties is overwhelming.

Since the method of superposed epochs has shown its potential to show the clear and also unambiguous response of SCIAMACHY NLC occurrence frequencies to the 27-day solar cycle, the analysis was extended to other NLC properties available from SCIAMACHY: radiance and altitude. Figure 8.10 presents examples of the response of the radiance at 291 nm and the NLC altitude. These time series were treated in the same way as the occurrence frequency. The graph shows that both NLC properties manifest a 27-day recurrence of the signal, and there is a marked 13.5 day component for the case of NLC altitudes. More important is the sign of the response to the 27-day solar forcing: while the radiance reacts similarly to the occurrence frequency, with lower radiance for higher solar activity, the NLC altitudes are actually correlated with the 27-day cycle. Both responses show a maximum response in
Figure 8.9: Superposed epoch analysis of the NLC occurrence frequency response to the 27-day solar forcing for SBUV data.
the vicinity of 0-day lag. The confidence level of the dependence between these variables and the Lyman-α irradiance exceeds 95%. It should be noted that the superposed epoch method was also employed on SCIAMACHY NLC particle size, but no significant response was observed.

Sensitivity

One desirable attribute of this type of analysis is the ability to isolate the mean response of the NLC properties to the 27-day cycle. This, in turn, enables the computation of the mean sensitivity of the affected variables to the forcing. While this calculation does not take into account the variation of the sensitivity to parameters other than the mean 27-day forcing, it is nevertheless a step further towards quantifying the impact of the 27-day cycle on NLC. The sensitivity is calculated through a simple linear regression of the mean NLC response and the mean Lyman-alpha forcing over a limited time range in the vicinity of the key event. The uncertainty of the calculated sensitivity is determined by fitting both signals with different phase lags and for different time periods. For instance, one can define the sensitivity of the rising edge of the Lyman-α, or simply for ±10 days around the key event.

The linear regressions for the determination of the sensitivity of NLC occurrence frequency and albedo measured by SBUV are shown in figure 8.11. The regressions presented are performed for data ±8 days around the key event, which maximizes the correlation coefficient. The correlation coefficients for the four cases presented, albedo and occurrence frequency in both hemispheres, are all larger than 0.97, yet another indication of the clear dependence of NLC properties on solar activity. The sensitivities calculated in this way are presented in table 8.2. Studies of the SBUV NLC properties sensitivity to the 11-year solar cycle have been carried out in the past by DeLand et al. (2003) and DeLand et al. (2007), among others. For comparison purposes, the results of these investigations are presented alongside the sensitivity of NLC properties to the 27-day cycle. It is remarkable that the
NLC albedo sensitivities to the 27-day and 11-year periodicities concur with each other within the uncertainty. This scaling of the sensitivity suggests that both phenomena could potentially originate from the same physical mechanism, assuming the process responds linearly to the Lyman-\(\alpha\) proxy. The 27-day sensitivity of the occurrence rate is, however, larger than that of the 11-year by about a factor of 2. As the occurrence frequency is not a direct physical value of the NLC, but a general measure of their activity confined between 0 and 1, it is not expected to respond linearly with the Lyman-\(\alpha\) irradiance.

SBUV data available for the present work were limited to the 60°-80° latitude band. This restriction however does not apply to the SCIAMACHY data set and therefore, a study of the dependence of NLC sensitivities on latitude can be undertaken. The sensitivity calculation of the SCIAMACHY NLC properties on the 27-day solar cycle is done in the same manner as for SBUV data. The data is partitioned in 5° bins, spanning the 50°-85° latitude range. Three different NLC properties are analyzed: occurrence frequency, albedo and altitude. The results are displayed in figure 8.12. In order to avoid confusion, the sensitivity will be discussed in terms of its absolute value, regardless of whether it is positive or negative.
Table 8.2: Sensitivity of the SBUV albedo and occurrence frequency to the 27-day and 11-year solar variation. Values of the sensitivities of NLC to the 11-year solar cycle were taken from DeLand et al. (2003) (occurrence) and DeLand et al. (2007) (albedo) for corresponding latitudes.

<table>
<thead>
<tr>
<th></th>
<th>Albedo</th>
<th>Occurrence</th>
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<tbody>
<tr>
<td></td>
<td>(-10^{-6} \text{ sr}^{-1})</td>
<td>(10^{11} \text{ photons cm}^{-2} \text{ s}^{-1})</td>
</tr>
<tr>
<td></td>
<td>27-day</td>
<td>11-year</td>
</tr>
<tr>
<td></td>
<td>27-day</td>
<td>11-year</td>
</tr>
<tr>
<td>NH</td>
<td>-0.61 ± 0.11</td>
<td>-0.706 ± 0.022</td>
</tr>
<tr>
<td>SH</td>
<td>-0.46 ± 0.12</td>
<td>-0.406 ± 0.026</td>
</tr>
</tbody>
</table>

A notable feature of the graph is that the sensitivity of all NLC properties is impacted by latitude. The NH occurrence rate is mostly unaffected by the 27-day cycle near 50°, but becomes increasingly sensitive to it with increasing latitude, reaching its largest sensitivity in the 60°-65° latitude bin. For subsequent latitudes, it weakens roughly linearly to reach a sensitivity value of \(-14 \%10^{-11} \text{ photons}^{-1}\text{cm}^2 \text{ s}\) near 85°, which is about half of its largest sensitivity. In the SH, the graph suggests that NLC occurrence rate response to the 27-day solar variation intensifies more or less linearly with latitude to attain its largest sensitivity, \(-13.5 \%10^{-11} \text{ photons}^{-1}\text{cm}^2 \text{ s}\), at the highest latitude.

For the NLC radiance, the sensitivity is largest at high latitudes and very weak for latitudes between 50°-60°. There is a threefold increase of the sensitivity between 72.5° and 77.5° in the NH, and more than twofold increase in the SH between 62.5° and 67.5°. The sensitivities for the NH and SH radiance differ roughly by a factor of 10, which is also similar to the ratio of their mean absolute radiance. Therefore, if scaled to the absolute radiance of the clouds, the sensitivity would be of the same order of magnitude.

Finally, the change in altitude linked to the 27-day solar cycle exhibits a peculiar behavior, with a very defined spike in sensitivity for both hemispheres. In the NH, the large sensitivity takes place in the 60° and 65° latitude band. The spike in the SH occurs between 65° and 70°, and its value is about the same as in the NH. For the last 3 latitude bands in both hemispheres, the sensitivity increases linearly, but at a moderate rate and to values about three times lower than that of the peak. The sensitivity of the altitude at lower latitudes could not be estimated due to the scarce number of NLC detections made at these latitudes and the sensitivity of the daily altitude to the number of detections. It should be mentioned that the amplitude of the altitude change (200-400 m) due to the 27-day solar rotation is within the uncertainty of the altitude retrieval (~600 m), and so would normally not be considered significant. Notwithstanding, the fact that the superposed epoch analysis showed a clear correlation with the Lyman-\(\alpha\) proxy and a 27-day recurrence was evidence enough to justify the calculation of the sensitivity.
Figure 8.12: Sensitivity of SCIAMACHY NLC occurrence frequency, radiance and altitude in both hemispheres on the 27-day solar cycle.
8.5 Discussion

The results from both SBUV and SCIAMACHY instruments show that there is strong evidence supporting the presence of a 27-day solar signature in NLC occurrence rates and that the signal is more clearly observed in the NH. As NLC can be affected by changes in temperature and H$_2$O, measurements from the MLS instrument of both of these parameters between 2004 and 2008 are analyzed in connection with the 27-day solar variation. A better understanding of the response of temperature and water vapor can provide clues as to the mechanism(s) responsible for the 27-day signature in NLC occurrence rates.

Water Vapor

Increased solar UV radiation is expected to lead to enhanced photolysis of water vapor in the upper mesosphere (von Zahn et al., 2004) and hence to a depletion of the H$_2$O molecules available for NLC formation, which could explain the observed anti-correlation of NLC occurrence with solar activity. Modeling of H$_2$O photolysis over the 11-year solar cycle shows depletion on the order of about 20% (1 ppm) at 83 km and 80% (0.8 ppm) at 90 km (Sonnemann and Grygalashvyly, 2005). There is also evidence that the ice particles also undergo photolysis, which could explain the vertical profiles of atomic oxygen in the vicinity of NLC (Murray and Plane, 2003, 2005). In order to verify the impact of the 27-day solar variability, mesospheric water vapor anomalies were calculated from MLS absolute water vapor mixing ratio, presented in figure 8.13. The contour plots of MLS H$_2$O mixing ratio anomalies (zonally averaged for the 60°-80° latitude band) vertical profile are shown in figure 8.14 for three entire NLC seasons and both hemispheres. The plots also present the Lyman-α anomalies over each season and the cross-correlation of both H$_2$O (at 85 km) and Lyman-α.

One of the main features of the contour plots is that the largest variability of H$_2$O in both hemispheres seems to be limited to altitudes above 80 km, with maximum amplitudes of 0.2-0.3 ppm at NLC heights which correspond to a variation of ~5% of the H$_2$O mixing ratio. This is consistent with the finding of Sonnemann and Grygalashvyly (2005), where only very small responses of H$_2$O to solar variability were observed below 80 km. The NH water vapor is positively correlated with the Lyman-α anomalies for year 2006 and 2007 with a phase lag of -1 and 0-day respectively. The NH summer season 2005 shows little or no correlation between H$_2$O and Lyman-α for 0-day phase lag and a negative correlation for a 5-day lag. In the SH, H$_2$O is not correlated with the Lyman-α anomalies for a phase lag of 0 day. According to these results, the upper mesospheric water vapor is not anti-correlated with the Lyman-α anomalies and is even sometimes positively correlated for a 0-day phase lag. Consequently, the water photolysis mechanism does not seem suitable to explain the 27-day modulation of NLC occurrence rate anomalies, since it happens usually with a 0-day phase lag, at least in the NH for the period covered by MLS data.
Figure 8.13: Vertical profiles of MLS water vapor mixing ratio and temperature during NLC seasons 2005-2008 in both hemispheres.

Figure 8.14: Vertical profiles of MLS water vapor mixing ratio anomalies during NLC seasons 2005-2007 a) in the NH and b) in the SH. Note that the color scale changes from year to year, as indicated by the legend above each contour plot. The solid line in the contour plots represents the Lyman-α anomalies over the seasons. The plots on the right hand side show the cross-correlation function of the Lyman-α anomalies and H₂O anomalies at 85 km, and the dashed line represents the correlation and anti-correlation statistically significant at the 90% confidence level.
Another way to investigate the effect of solar variation on H₂O is to apply the superposed epoch method to the data set during the summer season. Although only few years are available, the analysis has the potential of eliminating some of the noise which might be present. A complete analysis of the sensitivity of water vapor on the 27-day cycle for both hemispheres is presented in figure 8.15. The presence of a 27-day cycle is clearly seen in the upper summer mesosphere from the analysis of the Fourier spectrum of the superposed epoch response. Although the signal is present in both hemispheres, it extends to lower latitudes in the NH (∼55°N) than in the SH (∼65°S). It also extends to lower altitudes in the NH (∼70 km) than in the SH (∼80 km). An inspection of the absolute response of H₂O to the 27-day solar forcing shows that the amplitude is about the same in both hemispheres (∼0.2 ppmv peak-to-peak), but since the mean Lyman-α amplitude was lower in the SH, this leads to much larger sensitivity in the SH than in the NH. An analysis of the response lag shows that the maximum response is found about 7 days after the maximum in Lyman-α. Surprisingly though, the sign of the correlation is opposite in the different hemispheres. In the NH, there is a minimum in H₂O 7 days after a maximum in solar activity whereas in the SH, there is a maximum 7 days after the maximum in Lyman-α. Based on these results, the lag response of H₂O cannot explain the anti-correlation of NLC occurrence with solar activity, especially since the response is opposite in both hemispheres. It would be interesting to understand the reason for the difference of the sign of the correlation in the different hemispheres.
As the water vapor variation with the 27-day solar forcing is not consistent with the results from the analysis of NLC properties, especially concerning the phase lag of the response, the temperature data set from MLS is examined for a signal which could explain the observed NLC response. The temperature can be influenced by solar activity via a number of mechanisms in the upper mesosphere. Solar variation could affect atmospheric temperatures by changing the photolysis of O$_2$ through the reaction $\text{O}_2 \rightarrow \text{O} + \text{O}$, increasing atomic oxygen concentrations and chemically producing more ozone $\text{O}_2 + \text{O} \rightarrow \text{O}_3$. The enhanced solar irradiance and ozone concentration will increase the radiative heating rates by ozone as well as chemical heating rates. Temperatures could also be indirectly influenced by the solar variation, for instance through dynamical effects.

The absolute temperatures used for the calculation of the temperature anomalies are shown in figure 8.13 (right panel) for both hemispheres. Figure 8.16 shows MLS temperature anomaly profiles during three NLC seasons in both hemispheres, along with the Lyman-$\alpha$ anomalies and the cross-correlation of the solar activity proxy with temperature. For all NH seasons, there are significant correlations between temperature and Lyman-$\alpha$ anomaly for a 0-day phase lag. The correlation is especially good for years 2005 and 2006, seasons for which NLC occurrence rate anomalies are significantly anti-correlated with solar activity. In the SH, the situation is not as clear. Temperature and Lyman-$\alpha$ do correlate positively (although not significantly) for seasons 2004-2005 and 2005-2006, but show no correlation for season 2006-2007. These results are in good agreement with the NLC and Lyman-$\alpha$ cross-correlation analysis and suggest that solar radiation modulates mesospheric temperatures, which in turn affect NLC occurrence rates.

It is observe also that, while the amplitude of the temperature variation is about the same for all years, the 27-day solar UV forcing decreases from season to season. This is similar to what is observed in the NLC anomaly for both SCIAMACHY and SBUV. Gruzdev
et al. (2009) state that this effect is also present in their model simulations of the effect of the 27-day solar UV forcing on middle-atmospheric temperatures, where the sensitivities of temperature to solar activity generally decrease when the forcing increases. Another interesting conclusion of this model study is that, while there is a response in temperature to the 27-day solar cycle, it is intermittent and probably dependent on the dynamical state of the atmosphere. This can explain the absence of the signal in the NLC data set for some years.

The superposed epoch analysis is also applied to the temperature data set. The results are presented in figure 8.17. The analysis shows that for the NH, there is a clear correlation of temperature and Lyman-α around 0-day lag, with a peak-to-peak difference of about 1 K. The NH sensitivity of the temperature to the Lyman-α forcing is largest near the pole (∼4.5K \(10^{11}\) photons cm\(^{-2}\) s\(^{-1}\)) and decreases rapidly towards lower latitudes. The altitude of maximum sensitivity is found to be between 80 and 90 km for latitudes larger than 65° and drops to 65-75 km for latitudes between 45° and 65°. The sensitivity at low latitudes is not only found at lower altitudes but also to be lower in amplitude (1.5-2 \(10^{11}\) photons cm\(^{-2}\) s\(^{-1}\)). These results are in good general agreement with the SCIAMACHY sensitivity analysis. The radiance of NLC is most affected at latitudes polewards of 75°, where the sensitivity of temperature is clearly larger than at lower latitudes. The peak in the altitude sensitivity of NLC could be explained by the change in altitude of the maximum sensitivity, which crosses the bottom height of NLC at 65° latitude. The maximum sensitivity being directly at or near the bottom of the layer might affect more importantly the altitude at which particles sublimate and could explain the increase of 0.2-0.4 km observed for NLC altitudes between 60° and 65°. On the other hand, the radiance should also change accordingly, since the bottom altitude of the cloud affects the size up to which particles can grow, and particle size has been shown to affect the radiance. But since the differences are of the order of hundreds of meters, this might not be sufficient to notably affect the radiance. This is supported by the fact that no 27-day cycle could be observed in particle size, meaning that although it might exist, the effect is too slight to be significant.

In the SH, the response of temperature to solar forcing is strong in amplitude (∼2.4 K \(10^{11}\) photons cm\(^{-2}\) s\(^{-1}\)) and the 27-day component is clearly visible in the region between 70 and 90 km. However, the phase lag of the maximum response near 85 km is about 6 days. This is a much longer lag response than what was observed with the SBUV data set, but this is somewhat closer to what SCIAMACHY observed between 2003-2009, with a lag of about 4 days. It is important to keep in mind that the SBUV data used to infer the lag between the forcing and the response spanned decades while the MLS data set comprises only 4 years of measurements since late 2004. This might account for the discrepancy. Furthermore, the response of the temperature near the mesopause at 88 km shifts towards 0-day lag. It is conceivable that even though the sensitivity is smaller there than at lower altitudes, it affects the formation of NLC particles more dramatically. This result could explain the somewhat unclear phase lag response in the SH, since the NLC would be affected both during their initial growth phase near the mesopause and then later on for lower clouds. In any case,
there is a strong 27-day signal throughout the SH summer mesosphere, which extends from the pole to latitudes as low as 40°S. The sensitivity is maximum between 55°S and 75°S, and 75-85 km altitudes. Near the pole, the maximum sensitivity is closer to 85 km and sinks to 78 km around 70°S. As was the case for the NH, this decrease in maximum sensitivity altitude can possibly explain the peak in sensitivity of SH NLC altitudes near 70°S. The maximum sensitivity in SH NLC radiance is polewards of 65°S, and this could be interpreted by the fact that the maximum temperature sensitivity is found much closer to the SH NLC altitudes near the pole which are higher than the NH NLC altitudes.

Comparatively, the amplitude of the temperature variation in the SH is much larger than in the NH, leading to sensitivities which are about 3 times as large. On the one hand, it would be expected that due to this larger sensitivity, the NLC response would be clearer than in the NH, whilst in fact the opposite is the case. It is however well known that the NLC properties in the SH vary much more compared to their NH counterpart (Bailey et al., 2007). NLC in the SH are more affected by the interhemispheric coupling (Becker and Fritts, 2006; Karlsson et al., 2007) because stratospheric dynamics in NH is more unstable, and this may limit our ability to clearly observe the 27-day solar signature.
Concluding remarks

The examination of MLS data suggests that temperature changes in the upper mesosphere, not water vapor, are responsible for the observed variation of NLC properties on a 27-day timescale. There is much evidence (von Zahn et al., 2004; Sonnemann and Grygalashvyly, 2005) that water vapor will also be influenced by an increase in Lyman-α, and a 27-day signal is also observed in the data. H₂O variation in time however cannot explain the NLC observations. It is unclear whether the changes in water vapor are only due to a response to enhanced photolysis, or if the decrease in NLC activity for larger Lyman-α values, which would actually enhance the water vapor content by decreasing the ice content, can also account for part of the change in H₂O. Maybe this can explain the observed maximum in water vapor in phase with the minimum in NLC activity present in the SH. This, moreover, suggests that there are differences between both hemispheres, also in terms of sensitivity amplitude.

The presence of a 27-day cycle in temperature seems to explain the observed NLC properties variation with the solar activity. Other than the results presented in this chapter, a few references to a 27-day signal in mesospheric temperatures exist, albeit none concerning the summer mesopause region at high latitudes. Ebel et al. (1986) reported observations of temperature deviation of about 1.5 K at 80 km in the tropics, and argued that since the response to solar activity is mainly determined by the dynamical properties of the middle atmosphere, this means that the strongest perturbations should occur at middle and high latitudes. Model results of the 27-day UV forcing of the atmosphere from Zhu et al. (2003) agree well with this argument, showing an increasing sensitivity of temperature to the solar UV forcing with increasing latitude and altitude. Keating et al. (1987) also measured a 27-day signal in temperature in the mesosphere at low latitudes (±20°), but with a maximum sensitivity at 70 km. Although specific results of all these studies do not always concur, they are nevertheless consistent concerning the presence of a 27-day signal in middle-atmospheric temperatures which should affect the formation of NLC. The fact that the phase lag is about 0 day, in accordance with the NLC observations and a simple model from Brasseur et al. (1987), suggests a mechanism which quickly reacts with a change in solar variation. Direct solar heating, mainly by molecular oxygen, could explain the change in temperature in the upper mesosphere. Brasseur et al. (1987) also pointed out that dissociation of water vapor will also feed back to the temperature sensitivity in the upper mesosphere. Higher solar activity will produce OH through H₂O dissociation, which can oxidize O₃ and hence decrease the direct heating due to ozone. Model simulations are needed for a better understanding of these feedback mechanisms and their effect on temperature sensitivities.
Conclusions

The present work dealt with the retrieval of noctilucent cloud properties using SCIAMACHY limb measurements, and an analysis of the NLC response to the 27-day variation in solar activity. The study of NLC is of great interest for the determination and understanding of phenomena taking place near the summer mesopause.

First, the technique for the retrieval of NLC properties such as daily occurrence frequency, radiance, altitude and particle size was presented. Error associated with the retrieval of these parameters were also estimated. The particle size retrieval is based on the determination of the so-called Ångström exponent $\alpha$. The error on $\alpha$ is usually between 0.15 to 0.2, but is dependent on the brightness of the cloud. Even though the Ångström exponent can be retrieved in the SH, the particle size there can unfortunately not be determined because of the geometry of observation, since a single $\alpha$ value can lead to many different solutions. The assumptions made on the particle shape and PSD can be varied due to the retrieval based on look-up tables calculated using the T-matrix approach, offering a flexible retrieval scheme.

The compiled climatology of seven years of SCIAMACHY observations of NLC occurrence rate, radiance and altitude was presented. The NLC properties seasonal variations are in good agreement with other satellite and ground-based measurements. It has been shown that variation of NLC properties can be excellent indicators of atmospheric processes taking place at the summer mesopause, such as solar proton events, planetary waves (quasi 5-day...
wave) and gravity waves (zonal dependence of NLC occurrence frequency). Based on MLS data, it was shown that the NLC altitudes, despite the coarse vertical resolution of SCIAMACHY, correlate well with the level of supersaturation, especially in the NH. NLC mean altitudes in the NH lie between 82-83 km and are higher in the SH, near to 84 km. An inter-seasonal analysis of NLC properties does not show significant trends during the last seven years. No 11-year solar cycle is observed in the data set, which is in accordance with recent ground-based measurements from ALOMAR.

The first 7-year climatology of NLC particle size covering the entire NH with almost daily coverage has been presented. The seasonal variation of the NLC particle size is very similar to the seasonal variation of the occurrence rate and the radiance. It features, however, a large day-to-day variability. The latitude dependence of the particle size has been investigated and was shown to increase towards the pole. The magnitude of the variation depends on the PSD assumed. For a Normal PSD with $\sigma = 22$ nm, the increase of NLC particle with latitude is $\sim 1.5$ nm/$^\circ$. The anti-correlation of particle size with cloud height was also substantiated using different methods, leading to variation of the particle radius with heights on the order of 5nm/km and 15.3nm/km, the latter in accordance with different studies. The local time dependence of the particle size was also investigated and it was shown that for latitudes between 50-70$^\circ$ and local times of $\sim 11$ and $\sim 21$, the particle size difference was about 6 nm, with larger particle sizes found on the PM side of the orbit. The particle size computed throughout the last seven years seems to show a decreasing trend, especially marked during the last 2 years. This is surprising because one would expect that the reduced solar activity would be more favorable to large particle size formation. In addition, the sensitivity of particle size on the assumed particle shape and PSD was computed, showing large variations of the particle size depending on the retrieval assumptions. The change in PSD seems to affect the particle size more than the shape. The retrieved particle size is anti-correlated with the assumed PSD width as well as the axial ratio. Cooperation with other scientists retrieving NLC particle sizes using different experimental techniques led to interesting results during a particle size workshop. It was found that the most appropriate assumptions describing consistently all data sets was an NLC particle population following a PSD where $\sigma = r/2$ and with spheroids particles with AR=0.5. These results seem to confirm evidence of such particle size distribution from lidar measurements.

Finally, the last portion of the thesis focused on the effect of the 27-day variation in solar activity on NLC properties. Using both SCIAMACHY and SBUV data sets, a 27-day signal was discovered in occurrence frequency and albedo (radiance), statistically significant at the 99% confidence level. There is also a 27-day response found in SCIA altitudes, although the response is close to the uncertainty of the retrieved NLC altitude. There is evidence of the 27-day solar cycle in NLC properties using a correlation analysis with the Lyman-alpha proxy for different NLC seasons, but clear correlation between the different time series is only present for some years. The use of the superposed epoch method displays clearly the anti-correlation between NLC activity and occurrence rate (albedo) as well as a correlation of altitude with solar activity. The response time of NLC to the forcing is $\sim 0$-day, except
for SCIAMACHY SH data which shows only a weak anti-correlation between NLC properties in the SH and solar activity, and a lag of 5-day. This result is probably due to the short time series of SCIAMACHY and large natural variability taking place in the SH. Spectral analysis of the superposed epoch response even show features for periods of 13.5 days, further confirming that solar activity plays a role in the vicinity of the summer mesosphere. Sensitivity studies show that for SBUV data, the albedo response to solar forcing in both hemispheres is in agreement with the sensitivity of NLC to the 11-year solar cycle. The SBUV occurrence frequency sensitivity, however, is twice as large for the 27-day solar variation than for the 11-year solar cycle. Further analysis of SCIAMACHY data also features usually much stronger NLC properties sensitivity at high latitudes. An investigation of MLS temperatures and water vapor indicate that temperature seems to be the link between the change in NLC properties and solar activity, not water vapor. This is especially consistent with the 27-day signal observed in NLC altitude.

**Outlook**

In the future, more work could be done to enhance the NLC data set from SCIAMACHY. It has been recently pointed out by Jörg Gumbel that the strong spectral dependence in ozone absorption between 265-300 nm could affect significantly the Ångström exponent retrieved from NLC in limb geometry. Therefore, some modeling of the error introduced by neglecting this effect should be carried out. This should improve further the agreement with other particle size data sets.

The retrieval of NLC vertical ice mass column should also be implemented and compared with independent data sets, as this is a very interesting and physically representative quantity related to NLC activity.

It would be interesting to make use of the large data set of Ångström exponents in the SH. An analysis of their values could, if not permit the direct calculation of the particle size in the SH, at least lead to a set of constraints regarding NLC PSD and particle shapes which can explain the measured value of $\alpha$.

Finally, the feasibility of the retrieval of NLC in nadir geometry with SCIAMACHY should be assessed. If possible, a retrieval based on an algorithm similar to that of SBUV could be applied not only to SCIAMACHY, but also to similar instruments such as GOME and GOME II, and lead to a data set dating back to 1995.
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Bibliography


Baumgarten, G., J. Fiedler, and M. Rapp (2009), On microphysical processes of NLC: Observations and modelling of mean and width of the particle size-distribution, *ACPD (to be submitted).*

Becker, E., and C. von Savigny (2009), Dynamical heating of the polar summer mesopause induced by solar proton events, Submitted to J. Geophys. Res.


COSPAR (), The COSPAR International Reference Atmosphere (CIRA-86), Available from http://badc.nerc.ac.uk/data/cira/.


Ebisuzaki, W. (1997), A method to estimate the statistical significance of a correlation when the data are serially correlated, *J. Clim.*, 10(9), 2147–2153.


Rayleigh (1871), *Phil. Mag.*, 41.


von Zahn, U. (2003), Are noctilucent clouds truly a “miner’s canary” for global change, Eos, 84(28), 261.


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Erklärung

Hiermit erkläre ich, dass ich die Arbeit ohne unerlaubte fremde Hilfe angefertigt habe, dass ich weiterhin keine anderen als die angegebenen Quellen und Hilfsmittel benutzt und die den benutzten Werken wörtlich oder inhaltlich entnommenen Stellen als solche kenntlich gemacht habe.

Bremen, 07.Dezember 2009

Charles Robert