Stockholm University

Meteoric Aerosols in the Middle Atmosphere

Linda Megner

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Meteoric Aerosols in the Middle Atmosphere
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Linda Megner

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Stockholm University
Department of Meteorology
10691 Stockholm
Sweden

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Abstract

Meteoroids entering the Earth's atmosphere experience strong deceleration and ablate, whereupon the resulting material is believed to re-condense as smoke particles of nanometre size. These particles are thought to play a major role in middle atmospheric processes, effecting the charge and radiation balance, ice nucleation and chemistry. This thesis concerns the fate of the meteoric smoke in the Middle Atmosphere, and in particular its effect on the formation of ice phenomena such as noctilucent clouds (NLC), polar mesospheric summer echoes (PMSE) and polar stratospheric clouds (PSC).

The potential role of NLC as tracer for mesospheric processes and variability has generated substantial interest in these clouds. Once their formation is understood they can provide an important tool for monitoring this remote and inaccessible region. Today, it is well established that NLC consists of water ice, but the nucleation of ice in such a dry region has long puzzled the middle atmosphere research community. Supersaturation is not considered high enough for homogeneous nucleation to occur, thus pre-existing condensation nuclei are deemed necessary. Among the possible nuclei, smoke particles have long been considered the most likely. Generally, it has been believed that these particles exist in numbers of the order of thousands per cubic centimetre at the mesopause. This belief is based on global average studies of meteoric material that fail to take atmospheric circulation into account. Here we show that this circulation efficiently transports meteoric smoke particles away from the polar region before they coagulate large enough to efficiently act as ice condensation nuclei. This poses a challenge to our current understanding of mesospheric ice phenomena.

We show that the charging of meteoric smoke particles, in combination with deviations from the mean thermal state, may solve this dilemma by significantly altering the ice nucleation properties of smoke. Though there are measurements of charged particles in this region, the potential importance of charging processes has so far not been considered by mesospheric ice models. Although there is a large uncertainty in the fraction of smoke particles that are charged, we show that reasonable assumptions give number densities of charged condensation nuclei that are consistent with what is expected for mesospheric ice phenomena.

We further show that the bulk of the meteoric material is transported to the Arctic winter stratosphere, yielding significantly higher concentrations of meteoric smoke in the region of PSC formation than previously believed. Despite the intense research in PSC, their nucleation processes are not yet entirely understood and meteoric smoke has recently been argued a favourable condensation nuclei. Our new predictions of meteoric smoke in this region may thus shed new light on open questions related to PSC nucleation.

List of papers

This thesis consists of an introduction and the following five papers:

I Megner, L., Rapp, M., Gumbel, J., 2006: Distribution of meteoric smoke - sensitivity to microphysical properties and atmospheric conditions. *Atmos. Chem. Phys.* **6**, 4415-4426.

II Megner, L., Siskind, D. E., Rapp, M., Gumbel, J., 2008: Global and temporal distribution of meteoric smoke; a 2d simulation study. *J. Geophys. Res.* **113**, D03202, doi:10.1029/2007JD009054.

III Megner, L., Gumbel, J., Rapp, M., Siskind, D. E., 2008: Reduced meteoric smoke abundance at the summer pole - implications for mesospheric ice particle nucleation. *Advances of Space Research*, **41**, 41-49.

IV Gumbel, J., Megner, L., 2008: The importance of charging processes for mesospheric ice nucleation. *Submitted to Atmos. Chem. Phys.*

V Megner, L., Gumbel, J., 2008: Charged meteoric particles as condensation nuclei for mesospheric ice phenomena. *Manuscript*

Contents

Chapter 1

Introduction

Meteoric material is thought to play a key role in a number of middle atmospheric phenomena, such as noctilucent clouds (NLC), polar mesospheric summer echoes (PMSE), metal layers, charge balance and heterogeneous chemistry. As will be shown in this thesis, its influence is not restricted to the region where meteoroids ablate, but also effects processes in the underlying atmosphere and around the globe. Despite this obvious scientific interest, the fate of meteoric material in the atmosphere is not well known. The aim of this thesis is to gain better understanding both of how this material is distributed in the atmosphere and of the impact on middle atmospheric processes. In particular, the consequences for ice nucleation and formation of NLC, PMSE and polar stratospheric clouds (PSC) will be investigated. Interest in NLC is in particular due to their potential role as tracer for mesospheric processes and variability. Once the processes governing their formation are understood they can provide an important tool for monitoring this remote and inaccessible region. Lately there have been indications of an increase in the brightness and occurrence frequency of the NLC, while at the same time they seem to extend to lower latitudes. This has been suggested to be caused by anthropogenic forcing, and would thus be a first sign of climate change in the mesosphere. However, to understand if this is truly the case, better knowledge of the microphysics of NLC, such as nucleation and ice growth is required. Interest in PSC's is due to their key role in stratospheric ozone destruction. Massive effort has therefore been put into understanding their formation and interactions with the environment. Despite this, the nucleation processes of PSC are as of yet not entirely understood.

Although this thesis concentrates on the impact of meteoric material on ice particle formation, this topic is closely linked to a number of different topics; the circulation and mean state of the atmosphere, the influx of extraterrestrial material, the radiation and charge balance of the atmosphere and the microphysics of charging and particle growth. Before the results of this thesis are presented in papers I - V, some background about these topics is therefore given. Atmospheric modelling, which is the major tool for this study, is also discussed. Finally, the main conclusions from the publications are summarised and suggestions of how further knowledge can be gained are presented.

Chapter 2

Introduction to the Middle Atmosphere

In atmospheric science the atmosphere is traditionally decided into different altitude regions depending on how the temperature changes with height. The lowest region, the troposphere, reaches to an altitude between 7 and 17 km and is characterised by a decrease in temperature with height. This is the region which hosts our everyday weather and the majority of the clouds. At the top of the troposphere, the so-called tropopause, a temperature minimum of approximately -60°C is reached, above which the temperature starts rising again. This is mainly due to the ozone layer which absorbs ultraviolet sun light and thereby locally heats the atmosphere. This region is referred to as the stratosphere, and has gained attention lately because of the depletion of ozone. High aeroplanes fly in the lower stratosphere and unmanned balloons can reach the middle stratosphere. Above the ozone layer the temperature starts dropping again and we have reached the mesosphere. Only rockets can reach this remote region, making in-situ investigations very costly. In-situ observations of the mesosphere are therefore sparse comparing to the lower atmosphere, and even though satellite and lidar-observations have provided substantial information, there are many processes and phenomena that are not yet fully understood. One of these processes is the formation of ice particles at very low water concentrations, which this thesis deals with. At the top of the mesosphere, which is known as the mesopause, the temperature in the polar summer reaches below -140°C, making it the coldest place in the entire atmosphere. Above the mesopause, in the thermosphere, the temperature rises once again, this time due to absorption of high energy solar radiation by the the small amounts of oxygen and others species remaining at these altitudes.

The stratosphere and the mesosphere, which are the parts of the atmo-

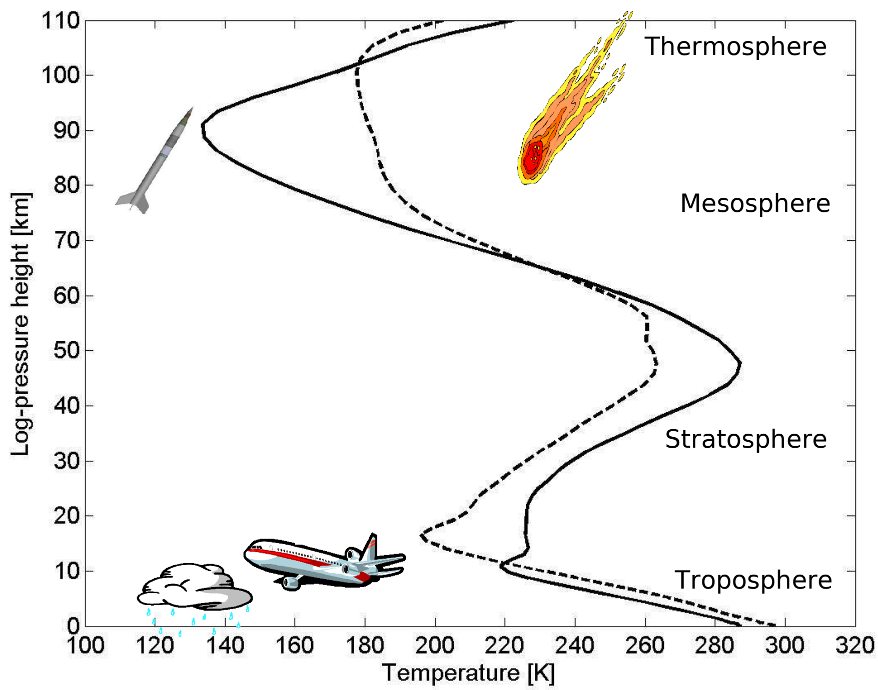

Figure 2.1: Schematic view of the temperature structure of the atmosphere and its division into various regions. The dashed line is a high latitude summer profile and the solid line represents the profile at the equator. From the Canadian Middle Atmosphere model (*Fomichev et al.*, 2002)

sphere this thesis is concerned with, are often commonly referred to as the "Middle Atmosphere". This region is different from the lower atmosphere in many ways; it is less dense, it is out of reach of convection so that air parcels do not regularly come in contact with the Earth's surface, the transport is driven in a different way (see next chapter), and it is much dryer. The reason for the latter is that most air enters this region in through the cold tropical tropopause, where the majority of the water is condensed, thus leaving air with water mixing ratio of the order of parts per million (ppm).

The exponential decrease of density with altitude results in very thin air which causes larger particles to rapidly sediment out of this region. The middle atmosphere can therefore only host very small particles for any length of time. Nevertheless, the influence of particles in the Middle Atmosphere is important. During the last decades it has been shown that particles influence both the charge balance and the chemistry of the Middle Atmosphere. As we shall see, they also enable ice formation in this very dry region, giving rise to the very high clouds and sporadic radar echoes described in chapter 4.

Chapter 3

General circulation of the Middle Atmosphere

In the atmosphere the zonal flows are generally the strongest. They are a direct consequence of the meridional temperature gradient as described by the thermal wind equation:

$$f\frac{\partial u}{\partial z} = -\frac{R}{AH}\frac{\partial T}{\partial \phi} \tag{3.1}$$

where u is the zonal wind velocity, z the "log-pressure" vertical coordinate [1], R the gas constant for air, A the radius of the Earth, H the scale height, T the temperature, f the Coriolis parameter and ϕ the latitude. On the Earth's surface the warmest temperatures are at the equator. For the northern hemisphere (NH) f is positive, so that the negative temperature gradient in latitude gives eastward zonal winds increasing with altitude. The positive temperature gradient of the southern hemisphere (SH) also results in vertically increasing eastward zonal winds, since f is negative in the SH. The meridional temperature gradient is thus the reason behind the eastward jets at approximately 11 km, see Figure 3.1. Further, the ozone absorption in the permanently sunlit summer polar stratosphere makes this the warmest region of the stratosphere, from where the temperature drops towards the winter pole. This temperature decrease with latitude gives rise to dominating westward winds in the summer stratosphere and eastward winds in the winter stratosphere as can be seen in Figure 3.1.

As mentioned in the introduction the circulation in the Middle Atmosphere is of different origin than in the lower atmosphere. The tropospheric

[1] The log-pressure coordinate z is used as an approximate altitude coordinate derived from the pressure according to $z = -H\ln(p/p_s)$ where p is the pressure and p_s is the surface pressure.

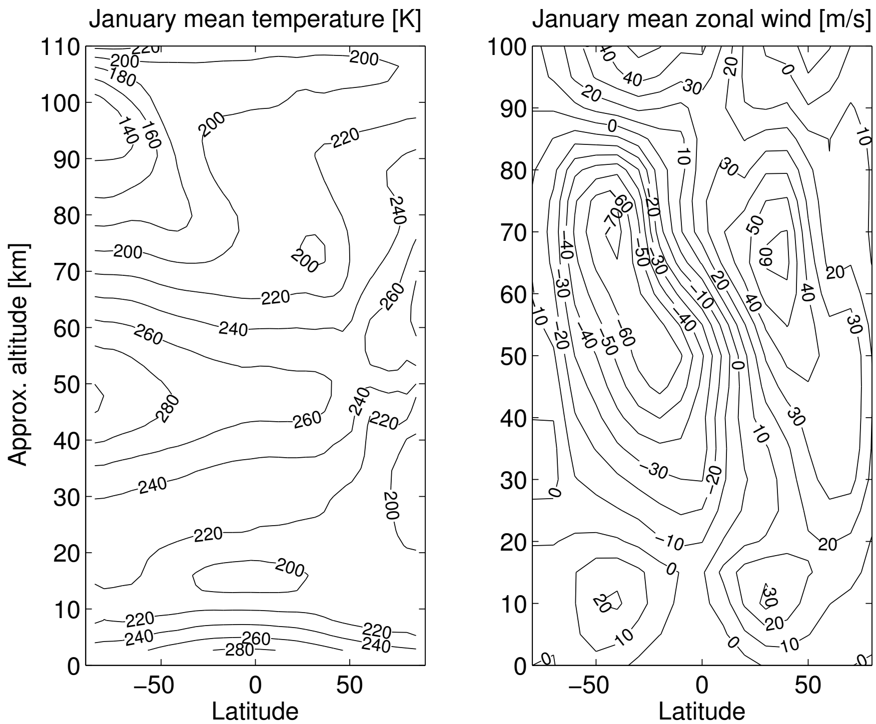

Figure 3.1: January temperature from the CHEM2D model (*Siskind et al.*, 1997, see paper II) and zonal wind field from the Cospar International Reference Atmosphere (CIRA).

circulation is, much like a classical heat engine, driven by the heat differences, which in turn result from uneven solar irradiation (more at the equator than at the poles) and uneven absorption of solar radiation (more at the ground than in the atmosphere). The high and low pressures systems, winds and weather are all consequences of this energy imbalance. In the Middle atmosphere the circulation is instead driven mechanically by deposition of angular momentum from breaking waves, originating from the troposphere. This wave momentum deposition is often referred to as "wave drag" and acts to disturb the radiative equilibrium. The waves responsible for the wave drag are Rossby waves (also known as planetary waves), which deposit their momentum in the stratosphere and gravity waves, which carry momentum up to the mesosphere. Stationary Rossby waves can only propagate in eastward background winds. In the stratosphere Rossby waves are thus restricted to the winter hemisphere, where, as we have seen, and eastward wind flow prevails both in the tropo- and stratosphere. These waves exert a negative wave drag as they break, i.e. decelerating the eastward motion, and thereby drive a poleward flow. Conservation of momentum then forces up-ward motion in the tropics and downward motion at the poles. The resulting flow cell is generally referred to as the Brewer-Dobson circulation, and exists as mentioned mainly in the winter hemisphere. The fact that it is cold air that rises at the tropical tropopause shows once again that the circulation is driven mechanically.

The gravity waves, which break in the mesosphere, can unlike the Rossby waves exert both positive and negative drag. However, the fact that they cannot propagate through a region where their phase speed is equal to the wind speed infers a filtering effect from the underlying atmosphere. The eastward winds in the winter stratosphere thus filters out gravity waves with positive phase speed so that mainly waves with negative phase speed reach the mesosphere (*Fritts and Alexander*, 2003). Negative phase speed gravity waves deposit negative momentum, i.e. they slow down the eastward flow, and drive a poleward circulation. In the summer hemisphere the situation is the opposite: The stratospheric westward wind flow allows mainly waves with positive phase speed through to the mesosphere where these waves deposit positive momenta and thus drive an equator-ward flow. Together the gravity wave drag from both the hemispheres thus drives motion from the summer to the winter pole. Here too mass conservation requires up-draft at the summer pole and downward motion at the winter pole (see Figure 3.2).

The down-welling over the winter pole adiabatically heats the winter polar stratosphere, as can be seen in Figure 2.1. The up-draft at the summer pole instead forces the air to cool adiabatically. The strength of this circulation is thus the reason why the coldest temperatures on the entire Earth is the summer polar mesopause (Figure 3.1). This mesospheric summer to winter circulation plays, as we shall see, an important role in this thesis. For a more detailed description of the circulation in the Middle Atmosphere see for instance *Shepherd* (2003).

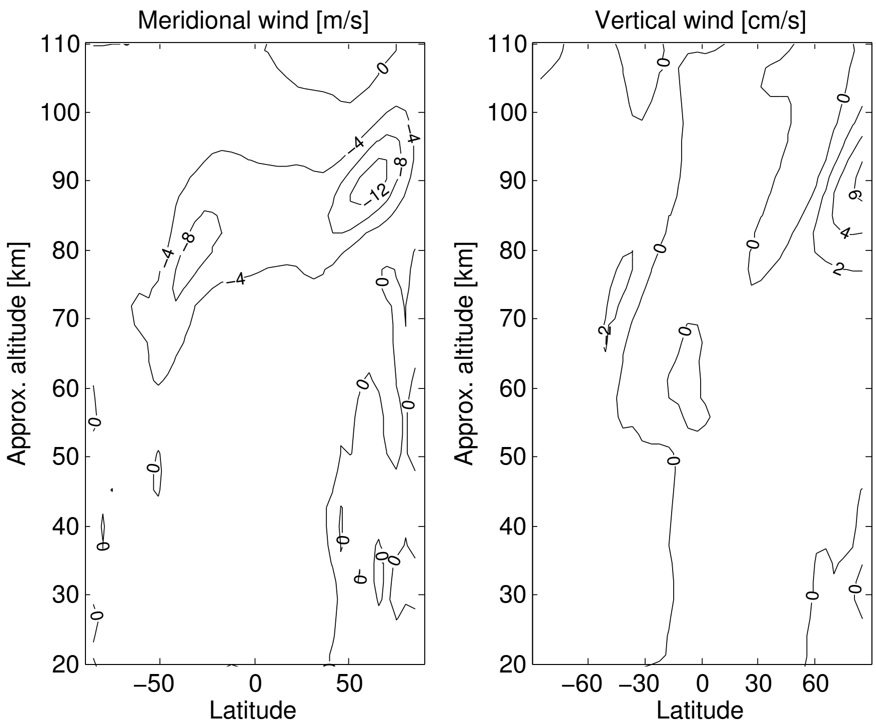

Figure 3.2: Meridional and vertical wind field from the CHEM2D model (see paper II) for January. The left panel show the meridional winds given in m/s, positive northward, and the right panel shows the vertical winds in cm/s, positive upward. The characteristic middle atmospheric circulation with a strong up-draft at the summer pole, transport towards the winter hemisphere and down-welling at the winter pole, is clearly seen.

Chapter 4

Middle atmospheric ice particle phenomena

4.1 Noctilucent clouds (NLC)

Noctilucent clouds (NLC), or Polar Mesospheric Clouds (PSC), are thin clouds appearing at altitudes of typically ∼83 km in the polar summer. When the sun disappears below the horizon but is still high enough to illuminate these high altitudes, the clouds can be seen as a lucent phenomenon - noctilucent clouds. The clouds consist of ice particles of typically 50 nm radius (*Witt*, 1968; *Thomas and McKay*, 1985; *Rusch et al.*, 1991, e.g.). The water content at the mesopause is measured in parts per million (ppm), as compared to parts per thousands where ordinary tropospheric clouds form. Very low temperatures are therefore needed to reach supersaturation. This is the reason why these clouds only occur at a very narrow altitude layer just below the cold summer mesopause. Even so, temperatures are not considered to be low enough to allow for homogeneous nucleation, i.e. spontaneous condensation of water vapour without a pre-existing nucleus. Given the limited input of material from the Earth to this region, it is not obvious what these nuclei could be. Candidates such as particles of meteoric origin (*Hunten et al.*, 1980), sulphate aerosols (*Mills et al.*, 2005), soot particles (*Pueschel et al.*, 2000) and ion clusters (*Witt*, 1969) have been suggested. Out of these, re-condensed meteoritic material, which this theses deals with, has long been considered the most likely.

Noctilucent clouds have been observed for over a century (*Leslie*, 1885). When their high altitudes were discovered, their mere existence proved that the atmosphere reached a lot higher than what was believed at the time. Still today, these clouds can tell us a lot about the mesosphere, such as water vapour and temperature trends, wind patterns and even about coupling

Figure 4.1: NLC display over Stockholm with tropospheric clouds in the foreground. (Courtesy of Jacek Stegman.)

Figure 4.2: PSC display over Esrange, northern Sweden. (Courtesy of Jacek Stegman.)

between the two hemispheres (*Karlsson et al.*, 2007). This is the reason why much research has been devoted to understanding their nature and properties (for a review see *Thomas*, 1991). As mentioned in the introduction there has lately been indications of an increase in the occurrence frequency and albedo of the NLC as well as a spread to lower latitudes (*DeLand et al.*, 2007). The lack of apparent temperature decrease at the summer mesopause (*Lübken*, 2000) suggests that the increase of NLC is due to an increase in water vapour, but changes in the number of nucleation seeds can not be ruled out. It has been suggested that these changes in NLC characteristics are the first sign of climate change in the mesosphere (*Thomas et al.*, 1989; *Thomas*, 1996), but in order to interpret what such an increase may mean, we first need to understand the mechanisms behind the formation of these clouds. One critical such mechanism is the nucleation process, which this thesis deals with. As we shall see this is linked to both the atmospheric circulation, gravity wave fluctuations, microphysical growth processes, radiation and charging processes.

4.2 Polar mesospheric summer echoes (PMSE)

Polar mesospheric echoes are radar echos that occur over a broad wavelength range ($\sim$20 cm - 100 m) close to the mesopause. They appear over the summer pole, hence the name, Polar Mesospheric Summer Echoes (PMSE). When first discovered (*Czechowsky et al.*, 1979; *Ecklund and Balsley*, 1981) the cause of the echoes was considered a mystery, but the following recent theory has proven successful in explaining their characteristics (*Rapp and Lübken*, 2004).

The same ice particles that form NLC are readily charged by thermal electrons. These particles are then transported by the turbulent velocity field so that structures in the distribution are created. The presence of these heavy negatively charged particles reduces the electron diffusivity and creates structures also in the electron distribution (*Kelley et al.*, 1987). Radar waves of the suitable wavelength undergo coherent scattering in these structures, yielding strong reflection in form of radar echoes. Because of the reduced electron diffusivity, the structures in the charge distribution can stay after the turbulence is gone which explains why PMSE can occur even when there is no turbulence. These echoes are observed over a broader altitude range than NLC, from approximately 80 to 90 km. This is because the ice particle generally grow as they sediment, and slightly larger particles are needed to scatter sun light and appear as visible NLC than are required to substantially reduce the electron diffusivity.

4.3 Polar stratospheric clouds (PSC)

Mother of Pearl clouds, formally known as Polar Stratospheric clouds, are extraordinary cloud formations that appear in the polar winter stratosphere at altitudes between 15 and 25 km (see Figure 2.1). Though beautiful in appearance, these clouds play a key role in the stratospheric ozone destruction. The existence of solid particles within a gaseous medium allows for chemical reactions that are not possible in a pure gas to occur on the surface of the particles - so called heterogenous chemistry. Heterogeneous processes on PSC particles activate chlorine compounds that initiate chain reactions leading to the destruction of ozone (for a review see *Solomon*, 1999). At the same time nitrogen compounds, which otherwise can de-activate chlorine species, are bound in a solid phase in the particle. Sedimentation of large such PSC particles leads to a permanent removal of nitrogen compounds (de-nitrification), thereby allowing the ozone destroying catalytic cycle to last longer.

PSC particles are, according to their main constituent, traditionally divided into liquid ternary solution droplets (STS), solid nitric acid trihydrate particles (NAT), and pure water ice particles. The limited water content of the stratosphere means that low temperatures are required to reach supersaturation and allow for condensation. These cold temperatures, $\sim$195 K or lower, only occur during the polar winter when the lack of sunlight prevents heating of the stratosphere from ozone absorption. In fact, even then the temperatures are on the border of being to warm, and the formation processes are not entirely understood. In particular, the formation mechanism for NAT is still under debate. The number densities of NAT particles appear too high to be explained by homogeneous nucleation of NAT (*Knopf et al.*, 2002; *Voigt et al.*, 2004). Nucleation on pre-existing ice has been suggested, but fails to explain all observed scenarios (*Carslaw et al.*, 2002; *Larsen et al.*, 2004; *Voigt et al.*, 2004).

Recently *Voigt et al.* (2004) suggested heterogeneous nucleation onto meteoric particles. As we shall see in this thesis, the expected concentration of meteoric aerosols in the PSC region is shown to be significantly higher than previously thought. Thus meteoric particles may play a more important role than previously believed (*Biermann et al.*, 1996).

Chapter 5

From falling stars to meteoric smoke

Falling stars, or meteors, have long fascinated people and played a large role in prophecy and myth. As the saying goes, a beloved child has many names, which is perhaps why these extraterrestrial rock phenomena are referred to by so many different terms. A meteoroid is an up to boulder-sized piece of debris in the solar system. As the meteoroid hits the Earth's atmosphere, it is heated by the intense deceleration, and ablates[1] causing a bright shine, which is known as a meteor. The very small meteoroids, typically smaller than 10 μm, have a large surface area relative to their mass, and can thus cool efficiently. These particles can therefore survive the impact of the atmosphere without ablating, and are referred to as micrometeorites. Larger meteoroids sometimes do not fully ablate, in which case the part that survives the atmospheric entry and can be found on the ground is known as a meteorite.

The mass influx of meteoroids to the Earth is believed to be dominated by the very largest objects, larger than 10^{13} kg, but these huge encounters only happen once every millions of years (*Ceplecha et al.*, 1998), and thus need not to be taken into account in this thesis. In fact, for atmospheric studies, we can neglect an even larger part of the mass influx, namely bodies large enough so that the majority of the mass survives the ablation ($\sim$1 m), and thus is not deposited in the atmosphere. By the same argument micrometeorites can also be neglected. As far as atmospheric studies are concerned, it is therefore enough to consider the mid-range of the meteoroid mass spectrum.

The surviving meteorites tells us what meteoric material is made of. Al-

[1]The word *ablation* refers to mass loss of any form and phase, such erosion, evaporation or melting. The process which causes the shine of the meteor is thus not burning, since this term normally denotes a process where oxidation occurs.

though it varies with the origin of the meteoroid, the dominating substances are Fe, Na, Mg, Si, Ca, Al and Ni (*Ceplecha et al.*, 1998). The grain density of meteorites typically varies from ∼2 g cm^{-3} to ∼5 g cm^{-3} depending on the type of meteorite (*Britt and Consolmagno*, 2003) . However, this thesis is concerned with the ablated material that is deposited in the atmosphere. This material is believed to re-condense to so called "meteoric smoke particles", which then are believed to play major roles in ice particle formation, surface chemistry and charge balance of the Middle Atmosphere. Even though these particles consist of meteoric material it is important to keep in mind that their properties may be very different from those of the incoming meteoroids.

Since 1961 when *Rosinski and Snow* (1961) first suggested the importance of re-condensed meteoric matter for the mesosphere, a number of attempts have been made to collect these particles (*Witt et al.*, 1963; *Kornblum*, 1970; *Gumbel et al.*, 2005). However, though there are many successful measurements of charged particles, presumably meteoric smoke, in this region (*Gelinas et al.*, 1998; *Horanyi et al.*, 2000; *Lynch et al.*, 2005; *Rapp et al.*, 2005, see Figure), there is as of yet no successful attempt to determine their structure and composition, or indeed even to ultimately verify their existence and meteoric origin. Thus there is lack of knowledge, not only about their properties, but also about their formation and interaction with the atmospheric environment. This lack of knowledge, combined with the limited knowledge about the mesosphere as a whole (comparing to the lower atmosphere), results in large uncertainties on the distribution and properties of meteoric smoke and thus on ice formation, surface chemistry and charge balance in the middle atmosphere.

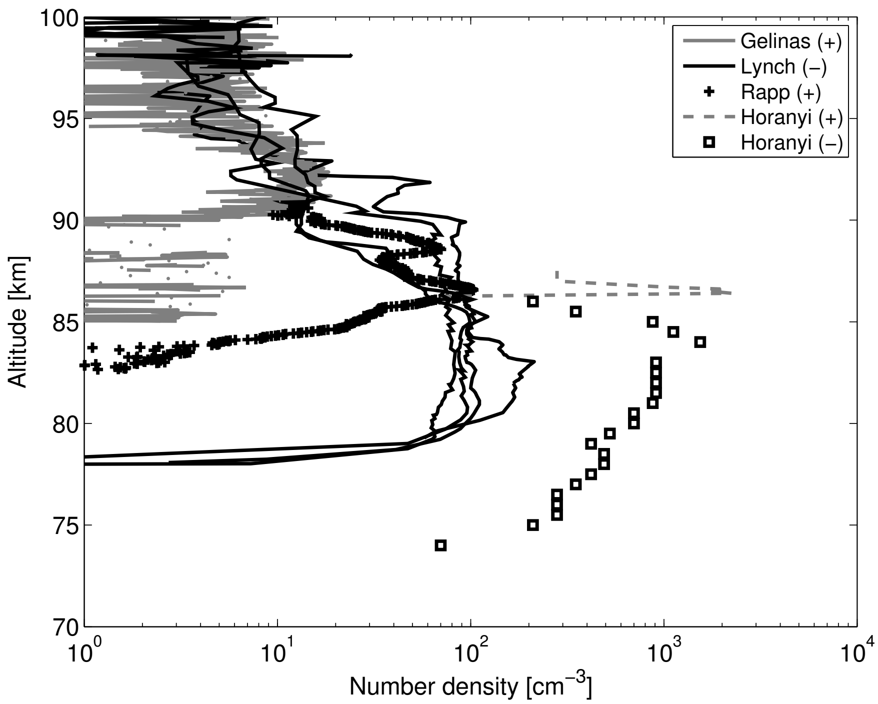

Figure 5.1: Measurements of number densities of heavy positive (+) and negative (-) charge carriers around the mesopause by rocket-borne particle impact detectors (*Gelinas et al.*, 1998; *Horanyi et al.*, 2000; *Lynch et al.*, 2005; *Rapp et al.*, 2005). These measurements will be discussed further in paper II.

Chapter 6

Atmospheric modelling

The atmosphere as a whole possesses an enormous number of degrees of freedom. In global weather forecast models for instance, the number of available observations is typically one order of magnitude less than the free parameters in the model. The atmospheric system thus is greatly under-determined. While this may seem obvious, it separates atmospheric science from many other experimental sciences where the aim is to restrict the amount of free parameters and recreate the same experiment enough times so that the system is overdetermined. This cannot be done in atmospheric science, both because our experiment (the atmosphere as a whole) is so large that we never can have the complete information about its state, and because we do not control it, and we can of course not re-run our experiment more than once. This is the reason why we construct computer models of the atmosphere, in which we do have control, and can re-run our experiment changing one parameter at the time. The lack of knowledge about the state is however still a problem. The atmosphere is chaotic, meaning that even a small difference between the model state and the state of the true atmosphere can grow to influence the whole system, what is in common talk sometimes referred to as the "butterfly effect". This is the basic reason why the weather forecasts occasionally can differ greatly from the observed weather. For this type of model the initial state of the atmosphere is of utterly importance since they are deterministic, i.e. their aim is to predict the state of the atmosphere at a certain time. Other types of models, such as for instance climate models and the models used in this thesis, use instead a mean state of the atmosphere and study how this mean state changes with time. The advantage is of course that the mean state can be determined more accurately than the state at any particular time. Another complication related to the largeness of the atmosphere is the limited amount of computation power. Even if we knew the exact state and location of every molecule, it would be impossible for a computer to handle all that information and compute every interaction in-between these molecules. This problem is handled by, instead of dealing with the molecules one by one, describing mean flows and mean chemical reaction rates. We thus calculate the mean velocity with which a certain volume of air moves and the mean rates at which its components are undergoing chemical reactions. The size of this volume is determined by the model resolution, and is also what determines the required computation power. In the state of the art global climate models it is of the order of 10 - 100 km. This is of course far too large to describe small scale processes, such as for instance turbulence, individual clouds or individual meteors. These small scale processes are therefore parameterised, i.e. described in a simplified way. As an example, molecular motion and small scale turbulence, which are both much smaller than the model resolution, are generally described using the diffusion equation, even though the nature of turbulence, especially as an effect of waves, is different to that of diffusion. It is therfore clear that parameterisations in general introduce errors. Moreover, it is difficult to determine the value of something like a diffusion coefficient for turbulence, simply because it cannot be measured, it is only a way to describe a small scale phenomena in large scale terms. The uncertainty limit in the vertical diffusion coefficient in the mesosphere region (10 - 1000 m^2/s, see paper I) illustrates this difficulty.

Atmospheric models can be 1, 2 or 3-dimensional where 1 dimension normally denotes the vertical direction and 2 dimensions normally refers to latitude-vertical. The disadvantage with less dimension is that processes that cannot be handled in the available dimensions must be parameterised, the advantage is of course less required computation power. The choice of model therefore becomes a trade-off between the required resolution, the region of the atmosphere that is to be modelled and the number of times we need to run the experiment, for instance with different parameter settings.

In this thesis modelling is used to investigate the fate of meteoric material in the atmosphere. To this end, processes of entirely different scales, reaching from the nucleation and growth processes of nanometre-size smoke and ice to the global transport of material by the atmospheric circulation, need to be represented.

The microphysics of growth of smoke particles and well as ice nucleation and growth is described by the Community Aerosol and Radiation Model for Atmospheres (CARMA) (*Turco et al.*, 1979; *Toon et al.*, 1979). The representation of growth of smoke particles and uncertainties arising from simplification of these processes are studied in paper I. The representation of ice nucleation is based liquid drop theory, which implicitly assumes that nanometre and sub-nanometre sized molecular clusters can be reasonably characterised using bulk properties of the condensed matter, possibly with some modifications to account for microphysical effects. Naturally, a rigorous microphysical description based on intermolecular interactions would be desirable, but is not feasible as quantum mechanical potentials and multi-body wavefunctions are not sufficiently well known. In lack of this, liquid drop theory is still the base of mesospheric ice models (*Hesstvedt*, 1961; *Turco et al.*, 1982; *Berger and von Zahn*, 2002; *Sugiyama*, 1994; *Rapp and Thomas*, 2006), and despite its limitations, it has proven to work surprisingly well in many circumstances (*Keesee*, 1989; *Pruppacher and Klett*, 1997). Its use needs to be judged against its ability to describe the experimental results.

In paper I the CARMA model was first used to vary all parameters of potential influence, in order to determine which of these parameters are the most important. Once it was shown that the wind field was of major importance it was clear that a global transport model also was needed. In paper II we thus couple the microphysics of the CARMA model to a 2-dimensional global chemical dynamical model, CHEM2D (*Siskind et al.*, 1997; *Summers et al.*, 1997; *Siskind et al.*, 2003), which transports the material according to the ideas discussed in chapter 3. The reason for choosing a 2-dimensional model, instead of a 3-dimensional, was to allow us to re-run the experiment many times in order to understand the importance of different processes.

Chapter 7

Major results from the publications in the thesis

As discussed in chapter 5 there is lack of knowledge about the composition and structure of meteoric smoke as well as a about its formation and interactions with the atmospheric environment. Paper I uses a one-dimensional microphysical model to investigate the sensitivities of meteoric smoke properties to uncertainties arising from this lack of knowledge as well as to variabilities in the surrounding atmosphere. Resulting uncertainty of meteoric smoke quantities such as number density, mass density, and size distribution are determined. The most important factor is found to be the background vertical wind and associated transport. The wind conditions in the polar summer mesosphere, where ice phenomena occur, is governed by a strong up-draft. The model indicated that the ablated meteoric material was swept away by this up-draft, leaving less than 1 potential condensation nucleus per cubic centimetre at the polar summer mesopause, which is far less than had been assumed earlier. However, the poor representation of transport in a 1-dimensional model made it difficult to judge the validity of these indications.

This spurred the development of a 2-dimensional model which could incorporate meteoric smoke and capture the seasonal and latitudinal variations of the vertical wind. This model, which consists of a coupling of a microphysical model and a global transport model, is described in paper II. The work confirmed the strong influence of the atmospheric circulation and showed that meteoric smoke particles efficiently are transported away from the summer mesopause before they have the time to grow large enough to serve as ice condensation nuclei. This contrasts the simplistic picture given by commonly used 1-dimensional meteoric smoke profiles, which refer to average global conditions, and yield of the order of a thousand nanometre sized particles per cubic centimetre at the mesopause, independent of latitude and time of year. At the same time the transport towards the winter pole and down into the polar vortex results in significantly higher concentrations of meteoric material in the winter stratosphere than previously thought. Our modelled number densities of particles are consistent with stratospheric balloon measurements of the so-called Condensation Nuclei (CN) layer. This is particularly interesting since meteoric smoke earlier, on the basis of the low concentrations of meteoric particles suggested by 1-dimensional models (*Zhao et al.*, 1995), has been disregarded as a cause of this layer. The enhanced concentrations of meteoric material in the winter vortex is of potential importance for stratospheric nucleation processes, which in turn effect PSC and ozone destruction.

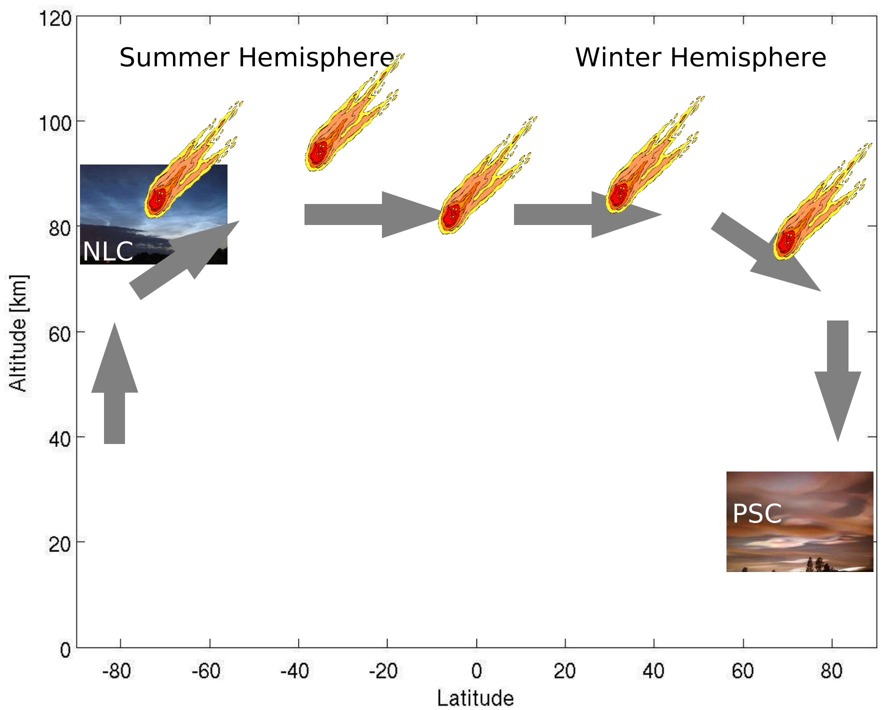

Figure 7.1: Schematic diagram showing how the atmospheric circulation transports the meteoric material from the summer polar mesopause, where NLC form, to the winter stratosphere, where PSC form.

In paper III we investigate the implications of low condensation nuclei number densities for the formation of ice phenomena. We find that even though resulting ice particle distribution may produce typical NLC brightness, the number density of ice particles is not consistent with what is expected for NLC and PMSE. In particular, it is two orders of magnitudes less than the concentration typically expected to explain the "electron bite-outs" that are frequently observed in the vicinity of PMSE's. We therefore re-examine the assumptions and parameters that determine the smoke distribution. We show that even though the number of condensation nuclei at the polar summer mesopause can be increased within the uncertainties, the results in most scenarios remain insufficient. We further suggest that small temperature deviations from the mean state of the mesopause, in combination with charging of particles, could solve this dilemma by significantly reducing the size limit above which smoke particles can act as ice condensation nuclei.

In paper IV ice nucleation on charged particles is examined. It is found that nucleation on charged smoke also is favourable because of another reason, namely that the charge attracts the dipole water molecules and thus the nucleus becomes more wettable. Paper IV also re-examines the concept of critical radius above which a neutral particle can act as condensation nucleus. This concept is based on the classical droplet theory, with the underlying assumption that molecular clusters can be reasonably characterised using bulk properties. This is, in lack of a more rigorous microphysical description, what mesospheric ice models are based on (*Hesstvedt*, 1961; *Turco et al.*, 1982; *Berger and von Zahn*, 2002; *Sugiyama*, 1994; *Rapp and Thomas*, 2006). It is found that the use of the critical radius as a parameter typically leads to a slight overestimation of the number of condensation nuclei, but is still a convenient way to characterise mesospheric nucleation conditions. Paper IV further states that it is not realistic to assume that a majority of all smoke particles are charged, since this would lead to large electron depletions throughout the mesosphere, which is not observed. This is a strong hint that the efficiency of meteoric smoke in capturing mesospheric charges must diminish towards smaller particle sizes. Our limited knowledge about charging efficiencies in the transition regime between molecular sizes and bulk particle sizes is today a major uncertainty for the understanding of mesospheric ice nucleation.

Paper V further examines charged mesospheric nucleation by analysing the time scales of processes such as charging, recombination, and particle growth. It is shown that positively charged particles, due to their rapid recombination with free electrons, under most circumstances cannot become effective ice condensation nuclei. Hence, for mesospheric nucleation it of primary importance to quantify the number of negatively charged particles. It is also shown that, under most atmospheric conditions, the smoke size distribution is surprisingly little influenced by charging effects on coagulation. This paper discusses the minimum size at which a smoke particle can accommodate the energy supplied by a captured electron, and thus form a stable charged particle, but a large uncertainty of this quantity remains. Also the large variability of ionospheric conditions implies a substantial uncertainty in the number of charged nuclei. Nevertheless, it can be shown that reasonable assumptions give number densities of charged condensation nuclei that are very consistent with what is expected for mesospheric ice phenomena. Thus, while it remains highly questionable whether neutral smoke can provide sufficient amounts of condensation nuclei for ice formation at the polar summer mesopause, charged meteoric smoke proves to be a promising candidate to explain mesospheric ice phenomena as we observe them.

Chapter 8

Outlook

As we have seen, our knowledge about basic parameters that determine meteoric smoke formation, such as composition, charge and shape of the particles as well as influx of extraterrestrial material, is still limited. This imposes considerable uncertainties on the ice nucleation properties of these particles. The main reason for the lack of knowledge is the complications involved in measurements at mesospheric altitudes where in situ studies only can be carried out using sounding rockets. Laboratory analysis of collected meteoric smoke is of primary importance to verify its meteoric origin and to determine its structure and composition. Laboratory studies of smoke charging efficiency, in particular for sub-nanometre clusters, are also needed in order to promote our understanding of mesospheric ice nucleation.

Much knowledge could be, and has already been, gained from measurement of number densities of heavy charge carriers in the mesosphere. However, the detection of nanometre-size particles using rockets is problematic since the shock wave in front of the rocket prevents small particles from reaching the detector. The size cutoff above which particles can be detected is a complicated function of instrument geometry, air density, and the rocket velocity. In order to be able to do meaningful comparisons of theory and data, experiments for which the aerodynamics can be understood and the size detection limit can be determined, are thus of great importance. Moreover, measurements of both neutral and charged particles, as well as simultaneous measurements of other atmospheric parameters, such as ionospheric conditions, chemical environment and temperature, are necessary to understand the distribution and nucleation properties of extraterrestrial material.

Complementary to in situ measurements, global measurements by satellite remote sensing would be highly desirable. However, the particles are too small for detection by scattered sunlight. A possibility is multi-wavelength satellite occultation experiments at longer wavelengths, which in principle can be used to determine the total smoke volume. Apart from providing a valuable tool for verifying model results, this could offer a unique opportunity to actually map, and even monitor, the global smoke distribution.

On the modelling side a more rigorous microphysical description of processes based on intermolecular interactions may be feasible, once the composition of smoke is better known. Ab initio molecular dynamics simulations can in principle teach us not only the possible structures of smoke clusters, but also about charge capturing and nucleation of of water onto such clusters.

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