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On the heterogeneous nucleation of mesospheric ice on meteoric smoke particles: Microphysical modeling

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ABSTRACT

Meteor smoke particles (MSP), which are thought to be the nucleation germs for mesospheric ice, are currently discussed to consist of highly absorbing materials such as magnesiowüstite, hematite or magnesium–iron-silicates and may therefore be warmer than the ambient atmosphere. In order to quantify the temperature difference between MSP and the atmosphere we developed a model to calculate the MSP equilibrium temperature in radiational and collisional balance. The temperature difference between MSP and the surrounding atmosphere strongly depends on the composition of the MSP, especially on the relative iron content, where a higher iron content leads to warmer MSP. We then derive an expression of the nucleation rate of mesospheric ice particles which explicitly accounts for this temperature difference. We find that the nucleation rate is strongly reduced by several orders of magnitude if the germ temperature is increased by only a few Kelvin. Implementing this nucleation rate depending on the germ temperature into CARMA, the Community Aerosol and Radiation Model for Atmospheres, we find that fewer but larger ice particles are formed compared to a reference scenario with no temperature difference between MSP and ambient atmosphere. This may indicate that iron-rich MSP are not ideal ice nuclei and that either other MSP-types or other nucleation pathways (e.g. wave induced heterogeneous nucleation or even homogeneous nucleation) are responsible for ice formation at the mesopause.

1. Introduction

The summer polar mesopause region is known for the coldest temperatures on Earth (~ 130 K Lübken and et al., 1990), low density and also as a part of the D- and lower E-region of the ionosphere. In combination with meteor smoke particles formed by re-condensing material of ablating meteoroids (Hunten et al., 1980) and neutral air dynamics some fascinating and scientifically interesting phenomena occur. These are, for example strong radar echoes, the so-called polar mesosphere summer echoes (PMSE) (Czechowsky et al., 1979; Ecklund and Balsley, 1981; Hoppe et al., 1988; Rottger et al., 1988) and the visible noctilucent clouds (NLC e.g., Jesse, 1896; Thomas, 1991). Both phenomena are closely related to the existence of ice particles in the mesosphere (e.g., Hervig et al., 2001, 2011; Rapp and Lübken, 2004). Even more than a century after their discovery the nucleation of these ice particles is still an ongoing topic in the current middle atmosphere research. It has been assumed by most earlier investigations that mesospheric ice particles form on the meteor smoke particles (MSP) by heterogeneous nucleation, although other nucleation mechanisms such as homogeneous nucleation (Murray and Jensen, 2010) or ion induced nucleation might also be possible (Witt, 1969; Gumbel, 2003). While the composition of MSP has still not been experimentally determined there are indications from lab experiments and satellite extinction measurements that meteor smoke particles are believed to consist of olivine ($Mg_{1.9}Fe_{0.1}SiO_4$), hematite (Fe_2O_3), different magnesium–iron-silicates ($Mg_xFe_{1-x}SiO_3$) (Saunders et al., 2010) or wüstite (FeO) and magnesiowüstite ($Mg_xFe_{1-x}O$) (Hervig et al., 2012). It is an obvious question how these different compositions influence the nucleation of mesospheric ice and which materials are more appropriate as nucleation germs or which can be excluded. Besides different surface properties the potential MSP materials differ in their radiative absorption coefficients, which leads to a material dependent MSP temperature. The current study seeks to quantify the abovementioned temperature difference between MSP and the ambient atmosphere and corresponding consequences for ice particle nucleation rates and polar mesospheric cloud (PMC)

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properties. In Section 2 we describe the model to determine the MSP equilibrium temperature as well as the equilibrium temperature of ice-smoke mixtures (i.e., “dirty ice”). In Section 3, we derive an expression for an MSP-temperature dependent nucleation rate which is followed by Section 4 in which results from such calculations are presented. Section 5 then finally demonstrates the effect of such modified nucleation rates on PMC-evolution using a full microphysical PMC-model. These results are discussed in Section 6 and our main conclusions are summarized in Section 7.

2. Equilibrium temperatures of MSP and dirty ice

2.1. Case 1: pure MSP

For the calculation of the equilibrium temperature of a spherical particle we assume that there are two power sources and two power sinks like former calculations from Fiocco et al. (1975), Eidhammer and Havnes (2001) and Espy and Jutt (2002). The balance between sources and sinks determines the equilibrium state of the particle. On one hand the radiation P_{sol} in the visible and ultra violet range of the sun and the terrestrial radiation P_{ter} in the infrared range are the power sources. On the other hand infrared radiation of the particle P_{rad} and collisions with air molecules P_{col} are the sinks of power. Further heating due to latent heat transfer of condensing water vapor will be neglected in this model. For steady state conditions all contributions to the power budget can be expressed by the following balance equation:

$$P_{sol}+P_{ter}-P_{rad}-P_{col}=0. \tag{1}$$

The first term P_{sol} of the balance equation (1) is defined as follows:

$$P_{sol}=\pi r^2\epsilon(1+2A\cos\chi)\int_0^\infty Q_{abs}(\lambda,r,n(\lambda))F_\lambda(T_\odot)\,d\lambda, \tag{2}$$

where r is the particle radius and $\epsilon=(R_\odot/R_0)^2$ with the sun radius $R_\odot$ and the sun–earth distance R_0 is the solar dilution factor which accounts for the small solid angle occupied by the sun. The factor $1+2A\cos\chi$ accounts for the direct radiation of the sun and the reflected radiation by the earth surface with albedo A at solar zenith angle χ. Q_{abs} is the absorption efficiency, $T_\odot$ the sun surface temperature and $F_\lambda(T_\odot)$ is the Planck formula for black body radiation

$$F_\lambda(T)=\frac{2\pi hc^2}{\lambda^5}\frac{1}{\exp\frac{hc}{\lambda k_B T}-1} \tag{3}$$

where c is the speed of light, h the Planck constant, λ the wavelength, k_B the Boltzmann constant and T the surface temperature of the radiating body. The absorption efficiency Q_{abs} weights the radiation function and depends on the radius and optical properties of the particle. Q_{abs} is calculated from the extinction and scattering efficiencies Q_{ext} and Q_{abs}

$$Q_{abs}=Q_{ext}-Q_{sca} \tag{4}$$

determined by the Mie scattering code *bhmie* of Bohren and Huffman (1983). Meinen et al. (2012) showed for the example of hematite (Fe_2O_3) that the bulk optical properties are still applicable down to particle diameters of ~2–3 nm. They found a good agreement between the experimentally determined extinction cross sections and those calculated by Mie theory.

The second term P_{ter} in the balance equation describes the particle heating by upwelling infrared radiation and is defined as

$$P_{ter}=\pi r^2\int_0^\infty Q_{abs}(\lambda,r,n(\lambda))F_\lambda(T_E)\,d\lambda. \tag{5}$$

The earth surface temperature T_E is assumed to be the temperature of the warm stratopause.

The particle itself radiates with its surface temperature T_P in the infrared. This power loss P_{rad} can be described as

$$P_{rad}=4\pi r^2\int_0^\infty Q_{abs}(\lambda,r,n(\lambda))F_\lambda(T_P)\,d\lambda \tag{6}$$

where we assume that the aerosols radiate homogeneously in all directions.

The collision between the particle and the air molecules can be an additional power loss term if the air is colder than the particle. Otherwise the particle will be warmed by the collisions with the ambient gas molecules. In free molecular flow energy is transferred by collisions from a unit area with a rate $\Delta\Phi^{(E)}$. The expression of $\Delta\Phi^{(E)}$ was taken from Gombosi (1994) and was also used in the calculations of Espy and Jutt (2002). We neglect the particle fall velocity and the horizontal winds since we assume that the MSP are moving with the background wind. With this assumption the original equation of Gombosi (1994) reduces to

$$\Delta\Phi^{(E)}=\frac{\alpha}{4}n_{gas}\bar{v}k_b\frac{\gamma+1}{2(\gamma-1)}(T_P-T_A). \tag{7}$$

The collisional energy transfer rate depends linearly on the thermal accommodation coefficient α which describes how efficient thermal energy is transferred between two bodies. T_P-T_A is the temperature difference between the particle and the surrounding atmosphere. n_{gas} is the surrounding gas number density with its mean thermal velocity $\bar{v}$. γ is the heat capacity ratio. The collision power P_{col} is obtained by integrating $\Delta\Phi^{(E)}$ over the aerosol surface. The particles are assumed to be spheres hence integration over the surface yields $4\pi r^2$:

$$P_{col}=4\pi r^2\Delta\Phi^{(E)}(T_A,T_P). \tag{8}$$

The particle temperature can now be derived from thermal equilibrium conditions described in Eq. (1). The power contributions of P_{rad} and P_{col} both depend on the particle temperature T_P, whereas P_{sol} and P_{ter} do not depend on T_P. However, P_{rad} is a nontrivial function of T_P so that a closed expression for T_P cannot be found. We therefore apply an iterative scheme, where the particle temperature of the previous step $i-1$ is used in the term of $P_{rad}(T_P^{i-1})$ to calculate T_P^i in the following manner:

$$T_P^i=T_A-\frac{P_{rad}(T_P^{i-1})-P_{sol}-P_{ter}}{\alpha\pi r^2 n_{gas}k_b\bar{v}\frac{\gamma+1}{2(\gamma-1)}}. \tag{9}$$

In the initial step the particle temperature is equal to the ambient atmospheric temperature ($T_P^0=T_A$). The iteration is terminated when T_P reaches an asymptotic value, i.e. $|T_P^i-T_P^{i-1}|<10^{-3}$ K.

The absorption efficiency Q_{abs} scales with r in the Rayleigh limit $r\ll\lambda$ which is fulfilled for MSP in the relevant wavelength range. All radiative power terms therefore scale with r^3, whereas the collisional loss term scales with r^2. Thus, larger particles will be dominated by the radiative terms and the comparably inefficient collisional loss will result in large particles being warmer than smaller ones.

Instead of assuming steady state conditions to derive the equilibrium temperature, the explicit time dependence of T_P can be solved for a given heat capacity of the particles (e.g. Chase et al., 1998 for hematite). We find that the time for a 2 nm hematite particle to reach equilibrium temperature is less than 0.1 s for conditions at 87 km.

2.2. Case 2: dirty ice – Maxwell–Garnett equation for effective dielectric permittivity

Mesospheric ice particles are believed to not only consist of ice but to include other materials e.g. the meteor smoke particles which were found by Hervig et al. (2012) with a smoke volume