Aerosol, Water and Clouds

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Hygroscopic growth and cloud formation

The interaction between airborne particles and water bare important implications several environmental issues, such as aerosol climate and health effects. Air usually contains much more water vapour (typically ~ g/m^3 in the atmosphere) than particulate matter (~ $\mu g/m^3$). Should only a small fraction of that water enter the particles, their physical and chemical properties would change dramatically. The phenomenon easiest to observe appears when there is a supersaturation with respect to water vapour, causing the formation of so called activated droplets. These droplets are not bound to an equilibrium state, implying that the droplets can grow to become large. This is what happens when clouds form in the atmosphere. Also with vapour concentration below saturation, particles are affected by water uptake. The relative humidity controls the size of dissolved particles by adjusting the vapour pressure at the droplet surface to reach the vapour pressure of the surrounding air by adding or removing water to the droplets. Our experience of condensation usually stems from macroscopic systems such as water in a jar, where evaporation to surrounding air is a slow process requiring days or more. When we are dealing with water uptake and evaporation in small systems like aerosol droplets, these processes are fast, for example a 1 µm diameter water droplet requires 1/1000 s to completely evaporate at low relative humidity.

1. Condensation and evaporation

A chemical substance is characterized by a saturation vapour pressure (p_0) that describes how much of the substance that can be in the gas phase. In a given system the gas phase concentration of the substance is described by the partial pressure (p). Its ratio to the saturation vapour pressure is called the saturation ratio (s):

$$s = \frac{p}{p_0} \qquad (1)$$

Over a plane surface of the substance the saturation ratio expresses the direction of the net vapour transport between gas and condensed phase:

s < 1(sub saturated):	evaporation
s = 1(saturated):	no net transport between the phases
s > 1(supersaturated):	condensation

Specifically for water there is a convention to express the saturation ratio as a percentage, which is known as the relative humidity (RH):

$$RH = 100s = 100\frac{p}{p_0} \qquad (2)$$

RH works in the same way as s with the value for saturation at RH = 100%.

The saturation vapour pressure of water, as well as for other substances, is strongly dependent on the temperature, see Figure 1. It can also be seen that the saturation vapour pressure is dependent on the phase, liquid or solid, of the substance. They coincide at the triple point with a

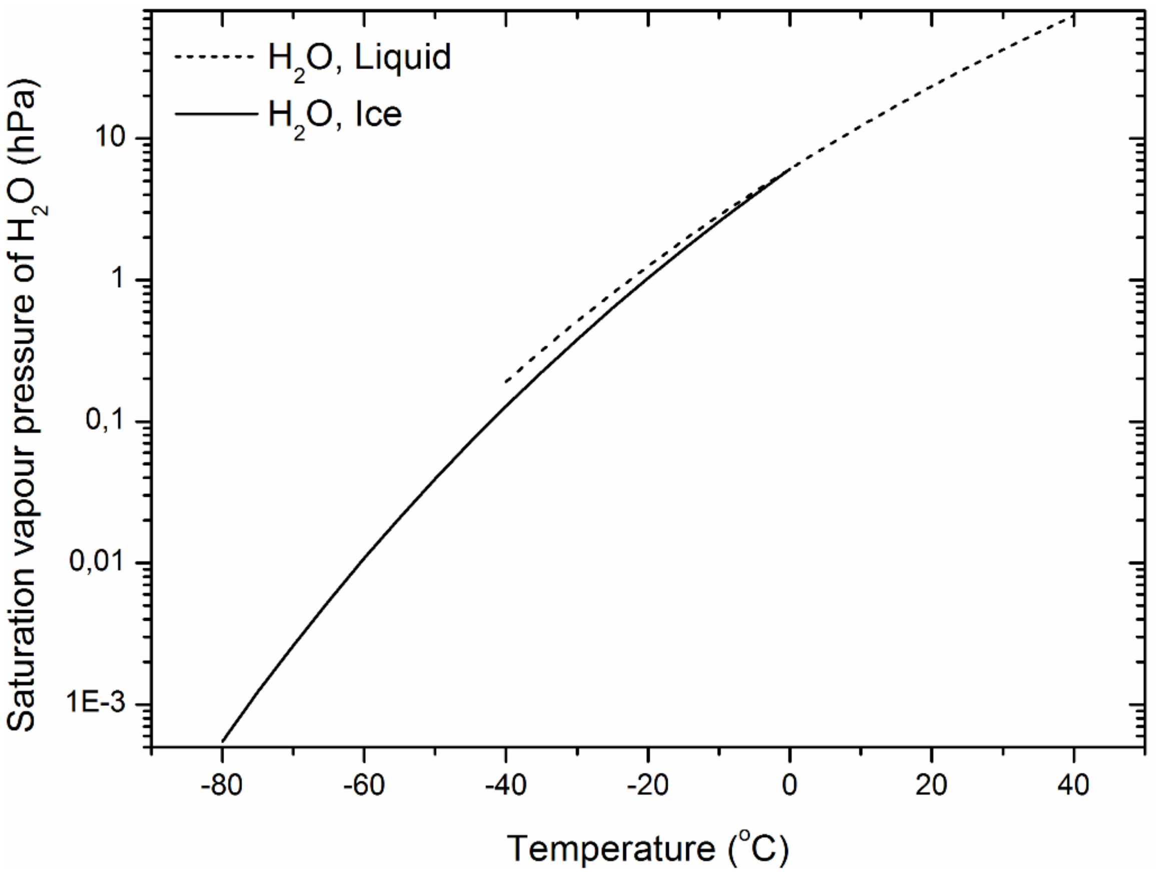


Figure 1. Saturation vapour pressure over liquid (-40 to +40º C) and solid (-80 to 0º C) water as a function of temperature.

growing difference as the temperature becomes lower. Later we will see that this difference between ice and liquid water has a strong influence on the formation of precipitation in the atmosphere.

It is thus not sufficient to know the partial pressure, we need also to know the temperature, and thereby the saturation vapour pressure, to describe a given system with respect to condensation and evaporation. Usually a graphic representation of the saturation vapour pressure does not provide sufficient precision when dealing with aerosol droplets. Instead tabulated values can be used. As an alternative, the saturation vapour pressure of ice and liquid water can be calculated from the modified Classius-Clapeyron equation:

$$p_0 = \exp(a_0 + a_1/T + a_2 \ln T) \tag{3}$$

Where T is the absolute temperature and p_0 is given in hPa. Valid ranges and values of constants a_i of equation 3 are given in Table 1 for ice and liquid water.

Table 1. Parameters and ranges of validity of equation 3 for liquid water and ice.

Phase	Range of validity	a_0	a_1	a_2
Liquid water	-40º C – +40º C	53.67957	-6743.769	-4.8451
Ice	-80º C – 0º C	23.33086	-6111.72784	0.15215

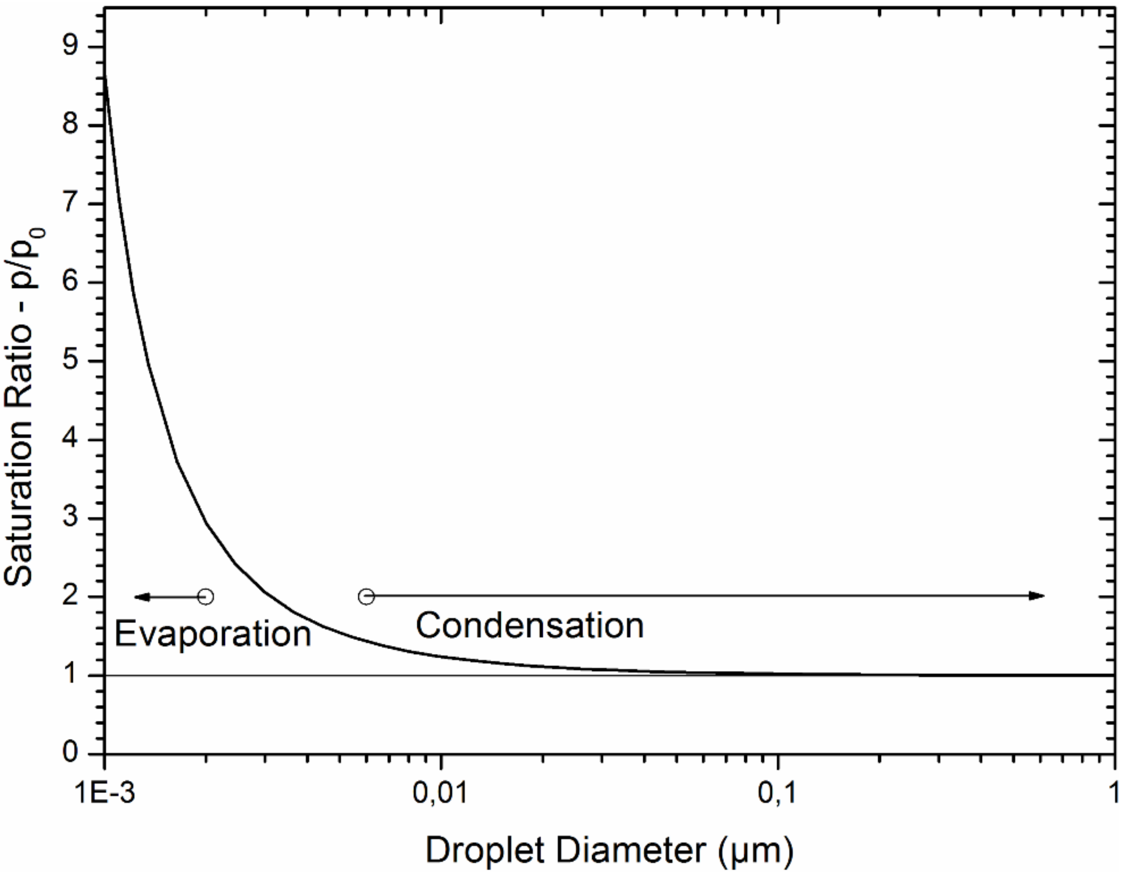


Figure 2. Limit between condensation and evaporation as a function of droplet size for pure water at 20º C.

2. The Kelvin effect

In the preceding section we discussed condensation and evaporation for a plane surface. When we want to study these phenomena for small systems, like airborne particles, we need to take into consideration that a droplet surface is curved. The curvature of the surface of a spherical droplet increases with decreased droplet radius. This modifies the attractive forces between the molecules at the surface. As a consequence the limit between net condensation and net evaporation is shifted to a higher partial pressure than for a plane surface. This effect, the Kelvin effect, is described by:

$$\frac{p_k}{p_0} = \exp\left(\frac{4\gamma M}{\rho R T d}\right) \tag{4}$$

where γ, M, ρ and d are the droplet surface tension, molar mass, density and diameter, and, R and T are the gas constant and the absolute temperature. A droplet of a given substance has the vapour pressure p_k at the surface, whereas that of a plane surface is p_0. For the partial pressure in the air surrounding a droplet of a pure substance we thus have:

$p < p_k$: the droplet evaporates

$p = p_k$: no net transport to/from the droplet
$p > p_k$: the droplet grows by condensation

From eqn. 4 we see that the Kelvin effect is dependent of droplet composition, and that the droplet diameter is in the denominator of the exponential, implying that the Kelvin effect decreases with increasing diameter. Figure 2 shows p/p_k for water at 20º C as a function of droplet diameter. It can be seen that the limit between condensation and evaporation strongly depends on the droplet size at small diameters, for large diameters the p_k/p_0 ratio approaches 1. This means that large droplets become more like a plane surface the larger they are, as a result of the reduced curvature.

3. Vapour pressure depression

We have this far discussed condensation and evaporation in pure substances. When a material is dissolved in the substance a reduction of the vapour pressure occurs, i.e. the limit between condensation and evaporation is shifted by the dissolved material. This vapour pressure depression becomes stronger with increasing concentration of the solute.

Let us now limit the discussion to a solution of water. For simplicity we also disregard the Kelvin effect in the following discussion; What happens with a water droplet containing dissolved material when the saturation ratio of the surrounding air is changed? Let us consider a droplet consisting a fixed amount of soluble material. Water vapour can be transported to and from such a droplet depending on the vapour pressure at its surface. Should this vapour pressure deviate from the vapour pressure of the surrounding air, this difference will drive a net vapour transport from the high to the low vapour pressure. The transport will continue until the vapour pressure of the droplet surface reach that of the surrounding air i.e. until the droplet reach equilibrium. This means that if the saturation ratio of the surrounding air is high, a small vapour pressure depression is needed. That is obtained by a low concentration of the solute. In this system the amount of solute is not changed, implying that the droplet has to take up much water to reach the low solute concentration and thus the same surface vapour pressure as the surrounding air. Applying the same reasoning for the case where the saturation ratio is low we find that we need a strong depression of the vapour pressure, implying that the solution need to be concentrated and therefore the droplet becomes small. In this way the relative humidity of the surrounding air controls the size of droplets at equilibrium.

Let us denote the relative vapour pressure depression Ω, i.e.

$$\Omega = p/p_0 \qquad (5)$$

This quantity is also known as water activity when used for water solutions. There is no general expression that can be applied to any kind of solution to calculate vapour pressure depression. The simplest kind of solutions are called ideal. The vapour pressure depression of an ideal solution is described by Raoult’s law:

$$\Omega_I = \frac{n_L}{n_L + j_I n_S} \qquad (6)$$

where n_L and n_S are the mol amounts of solution and solute. Many substances are dissociated in solution. j_I describes the number of dissociation products, for example NaCl forms Na^+ and Cl^- in solution implying that $j_I = 2$. From eqn. 6 it can be seen that $\Omega_I = 1$ for the pure solution (i.e. when $n_S = 0$) and that är Ω_I is less than 1 when $n_S \neq 0$. For water solutions the requirements of Raoult's law are approximately fulfilled when the solute concentration is low. For dissolved airborne particles this is approximately obtained for relative humidities close to 100%. For this kind of droplets it is common to express Raoult's law:

$$\Omega_I \approx \left(1 + \frac{6 j_I m_S M_w}{M_s \rho_w \pi d^3}\right)^{-1} \qquad (7)$$

where d, ρ_w and M_w, are droplet diameter, water density and molar mass, whereas m_S and M_s are mass and molar mass of the solute.

Raoult's law is invalid for a large number of solutions. It is beyond the scope of this book to give a comprehensive presentation of non-ideal solutions. Here the van't Hoff factor (j_N) is presented as an example of methodology. Resulting equation is similar to Raoult's law, where the factor j_N replaces j_I:

$$\Omega_N = \frac{n_L}{n_L + j_N n_S} \qquad (8)$$

The factor j_N describes the deviation from the ideal solution. Sometimes it is seen as the degree of dissociation of the solute, which would then indicate that j_N should always be smaller or equal to j_I. This is however not the case. Besides degree of dissociation other deviation from the ideal solution can appear. For example NaCl in a water solution j_N in some concentration ranges exceeds j_I (j_I of NaCl is 2). To deal with non-ideal solutions specialized literature need to be consulted to find relations between the solute concentration and relative vapour pressure depression. We illustrate the non-ideal solutions with the following exercise:

> ***Exercise 1:*** The van't Hoff factor for an important component of the atmospheric aerosol, ammonium sulphate ($(NH_4)_2SO_4$), can be calculated from:
>
> $$j_N = 1.9242 - 0.1844 \ln C - 0.007931 (\ln C)^2 \qquad C \leq 1$$
>
> Where C is the molality of the solution (mole solute per kg of solvent)
>
> a) Calculate the van't Hoff factor and the relative vapour pressure depression for solutions with C = 1 mole/kg and 5×10^{-5} moles/kg.
> b) Calculate the molalities for an ideal solution for the in (a) calculated relative vapour pressure depressions
> c) Conclude on the differences between ideal and non-ideal treatment of the solutions
>
> *Answer a)* The van't Hoff factor is calculated from the given equation. Knowing j_N we can calculate the relative vapour pressure depression by first identifying

$$C = \frac{n_S}{m_w} = \frac{1}{M_w}\frac{n_S}{n_W} \qquad \Rightarrow \frac{n_S}{n_W} = CM_w$$

Which can be used in equation 8

$$\Omega_N = \frac{n_W}{n_W + j_N n_S} = \frac{1}{1 + j_N\, n_S / n_W} = \frac{1}{1 + j_N C M_w}$$

Entering j_N and C gives the sought vapour pressure depressions. Results:

C (mol/kg)	j_N	Ω_N
1	1.924	0.966
5 10^{-5}	2.973	0.999997

Answer b) We use equation 6 for an ideal solution. For ammonium sulphate $j_I = 3$, because the salt can dissociate in two ammonium an one sulphate ion. In the same way as in (a) we re-arrange in the Ω_I equation to extract an expression for molality assuming ideal solution (C_I):

$$C_I = \frac{1}{j_I M_w}\left(\frac{1}{\Omega_I} - 1\right)$$

Ω_I takes the values calculated for Ω_N in (a):

$C_I(0.966) = 0.641$ moles/kg; $C_I(0.999997) = 4.95\ 10^{-5}$ moles/kg

Answer c) In order to compare the ideal to the non-ideal calculations we form the ratio C_I/C. We obtain C_I/C 0.64 and 0.99 for the relative vapour pressure depressions 0.966 and 0.999997. Should we measure the relative vapour pressure depression of the former solution and assume ideal solution to compute the concentration of ammonium sulphate, we would underestimate the concentration by 36%, whereas we would be close to the correct value for the diluted solution.

4. The Köhler equation

In the two preceding sections two phenomena with strong influence on the interaction between water and airborne particles have been described. The effects of the curvature of the surface of droplets and vapour pressure depression acts together in droplets. Köhler was the first to deduce an expression that combines these two effects. The original expression is valid for diluted droplets consisting of ideal solutions:

$$\frac{p}{p_0} = \Omega_I \exp\left(\frac{4\gamma M}{\rho R T d}\right) \approx \left(1 + \frac{6 j_I m_S M_w}{M_s \rho_w \pi d^3}\right)^{-1} \exp\left(\frac{4\gamma M_w}{\rho_w R T d}\right) \qquad (9)$$

The Köhler equation connects droplet size (d) with saturation ratio (p/p_0) for a given particle (i.e. the material dissolved in the droplet) described by mass (m_S), molar mass (M_S) and number of dissociation products (j_I). Figure 3 shows the Köhler equation applied to an NaCl particle that has the diameter 0.05 µm when completely dry. We see that the curve has a characteristic

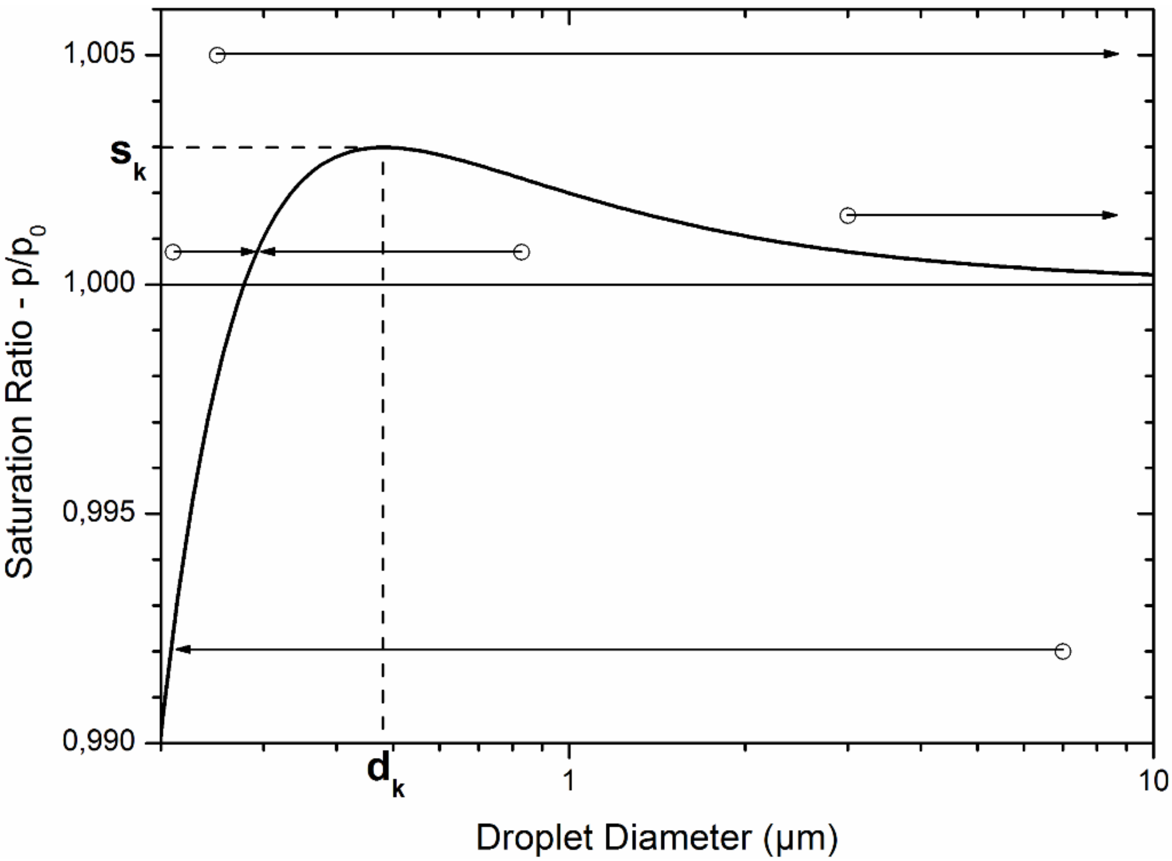


Figure 3. The Köhler equation applied to a droplet formed on an 0.05 µm diameter NaCl particle at 20º C, where s_k and d_k are the critical saturation ratio and the critical diameter. The circles and arrows illustrate regions of equilibrium and droplet activation.

maximum, the critical saturation ratio (s_k) which is reached at the critical droplet diameter (d_k). It can be seen that two droplet diameters are obtained in the saturation ratio range of $1 < s < s_k$. However, these two droplet sizes have different meanings. When $d > d_k$ the Köhler equation in the same way as the Kelvin equation describes the instable limit between condensation and evaporation, the section of the Köhler equation for $d < d_k$ describes stable equilibrium sizes of droplets as a function of saturation ratio. When the critical saturation ratios is exceeded, droplets are no longer bound by equilibrium. They can grow beyond the critical diameter and form activated droplets. This is what happens when clouds form. The droplets can in principle grow to very large sizes. In practice, availability of water vapour limits the growth to 20 – 30 µm diameter. We thus identify two kinds of droplets, those that are activated and those who are bound by equilibrium. The range where droplets are bound by equilibrium is given by:

- $d < d_k$: bound when $s < s_k$
- $d > d_k$: bound when s is smaller than the value calculated from the Köhler equation

For saturation ratios outside this range droplets are activated. The marked examples (open circles) in Figure 3 illustrates by the arrows which droplets are bound by equilibrium and which that are activated.

Using equation 9 to calculate d_k and s_k is rather time consuming. A common approximation is based on a Taylor approximation of the exponential term and an approximation of the relative vapour pressure depression for diluted solutions ($n_S/(n_L+n_S) \approx n_S/n_L$). For saturation ratios close to 1 the Köhler equation can be approximated by:

$$\frac{p}{p_0} \approx 1 - \frac{6 j_I m_S M_w}{M_s \rho_w \pi d^3} + \frac{4\gamma M_w}{\rho_w R T d} = 1 - \frac{c_1}{d^3} + \frac{c_2}{d} \qquad 10$$

This approximation for ideal solutions can be used to estimate the critical parameters. By forming the derivative by d of equation 10, d_k is obtained, and s_k is obtained by entering d_k in 10:

$$d_k = \sqrt{\frac{3c_1}{c_2}}; \qquad s_k = 1 + \sqrt{\frac{4c_2^3}{27c_1}} \qquad 11$$

where c_1 and c_2 are defined in equation 10.

> ***Exercise 2:*** Estimate the critical saturation ratio and droplet diameter for the particle in Figure 3, i.e. a NaCl particle having diameter 0.05 µm that has been dissolved in water. The temperature is 20° C and the surface tension 0.073 N7m.
>
> *Answer:* We calculate the values of c_1 and c_2
>
> $$c_1 = \frac{6 j_I m_S M_w}{M_s \rho_w \pi}; \qquad c_2 = \frac{4\gamma M_w}{\rho_w R T}$$
>
> and enter in equation 11. We obtain $d_k = 0.482$ µm and $s_k = 1.002992$. When we use equation 9 the results are $d_k = 0.481$ µm and $s_k = 1.002987$. The approximation thus worked well in this case. For smaller particles the deviation of the approximation increases.

The Köhler equation in the form of equation 9 is not applicable for many situations. In the previous section we saw that j_I need to be replaced by the concentration-dependent van't Hoff factor for concentrated solutions. It is, however, not possible to replace j_I in equation 9 with a concentration-dependent j_N because this equation was deduced using that the derivative by the solute concentration of j_I is 0. Additional corrections concerning dependences on the concentration of droplet density and surface tension are needed. The resulting expression for non-ideal solutions is complicated and beyond the scope of this book.

Although the concentrated solutions is not described quantitatively, a qualitative description will be given. Water soluble substances can be truly hygroscopic or deliquescent. The former compounds are characterized by water uptake at all saturation ratios and increased uptake with increased humidity. Deliquescent compounds has no water uptake at low saturation ratio. When the humidity is low, but increasing the deliquescent particle remains dry until the saturation ratio reaches a value characteristic of the compound where the dry particle dissolves and forms saturated solution droplet, see Figure 4. For e.g. NaCl this characteristic saturation ratio of droplet formation is 0.76. When the humidity is further increased the droplet grows by water

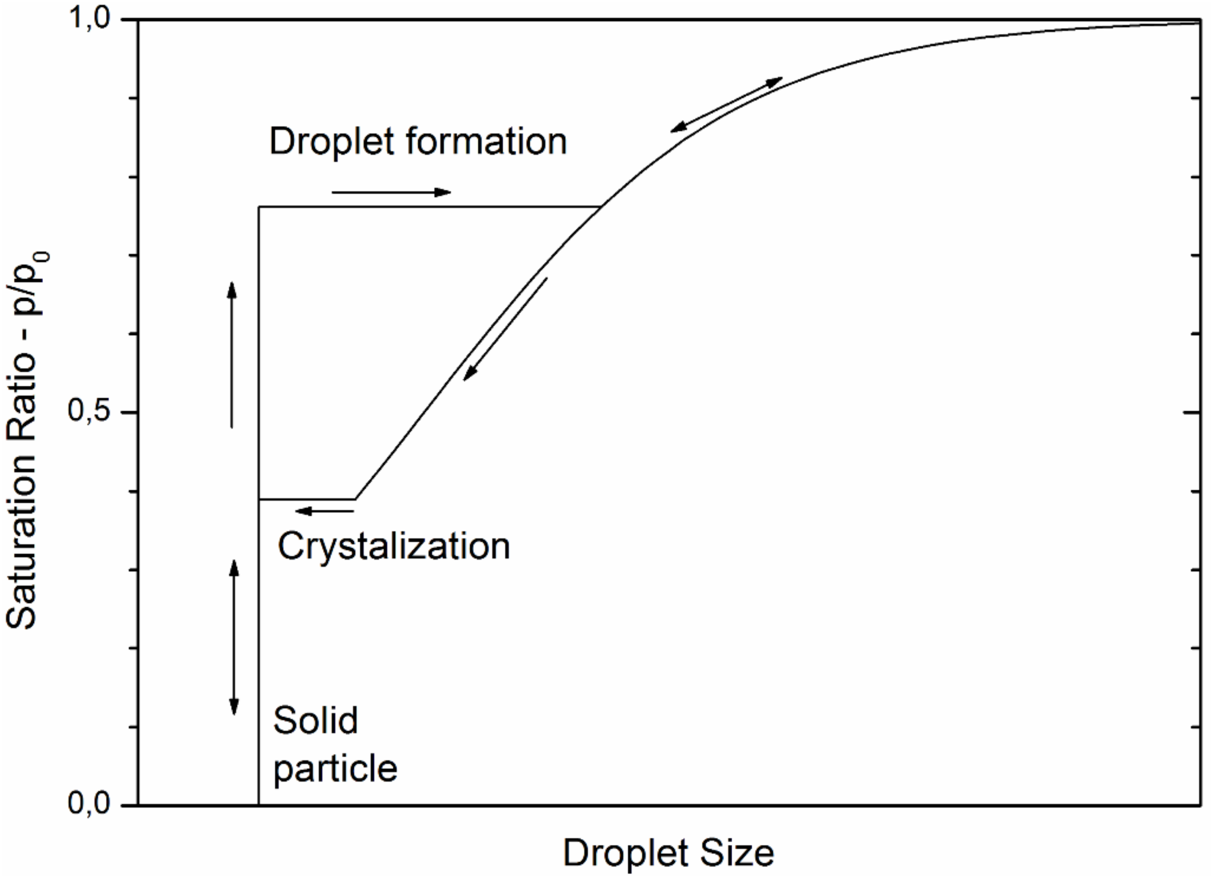


Figure 4. Droplet formation and crystallization for particles consisting of a deliquescent compound. The arrows indicate where the saturation ratio increase or decrease.

uptake, i.e. the droplet vapour pressure depression adjusts to the vapour pressure of the surrounding air. Turning the process around to decreasing humidity, the droplet size decreases. When the deliquescence point is passed the droplet continues to shrink, forming a supersaturated solution droplet. The shrinkage continues until saturation ratio of 0.3 to 0.4 is reached, before the droplet crystallizes. This hysteresis effect with droplet formation and crystallization at different saturation ratios is a regular feature of small droplets containing a deliquescent solute, whereas uncommon in large systems such as a solution in a jar.

This far we have limited our discussion to water solutions containing a single solute. When two deliquescent solutes are mixed, the two substances interact in a way that droplet formation appear at other saturations ratios than for the pure compounds. A mixture of several deliquescent compounds behave similarly to truly hygroscopic compounds in that they take up water also at low humidities and shows no clear steps in size from droplet formation.

Organic compounds attracts much attention in scientific research on the water uptake by atmospheric aerosol. These compounds co-exist in atmospheric aerosol particles with inorganic, water soluble substances like sulphates and nitrates. Organic substances are a large group of atmospheric compounds exhibiting a large variety of properties. In different ways they can affect the water uptake of aerosol particles and cloud formation. Their water solubility vary in a wide range. Limited soluble organic compounds do not appreciably affect particle water uptake at low humidities. When the droplet grows in size at relative humidity close to 100% these compounds can enter the solution and that way affect the critical saturation ratio, thus contributing to cloud droplet formation. Droplet activation can also be affected by surface active organic compounds, i.e. molecules containing both hydrophilic and lipophilic parts. These compounds tend to be distributed over the droplet surface rather than in its interior, affecting the surface tension thereby lowering the Kelvin effect and thus the critical saturation ratio. Insoluble chemical compounds do not directly contribute to water uptake. However, when the insoluble part is a large fraction of a particle, the size and thus the Kelvin effect of the droplet formed can be affected by the volume of the insoluble inclusion. We conclude by presenting the Köhler equation of an ideal water solution formed on a particle containing one insoluble and one soluble part, with the soluble volume fraction ε':

$$\frac{p}{p_0} \approx \left(1 + \frac{6 j_I m_S M_w}{M_s \rho_w \pi (d^3 - (1-\varepsilon') d_{dry}^3)}\right)^{-1} \exp\left(\frac{4\gamma M_w}{\rho_w R T d}\right) \qquad (12)$$

where d_{dry} is the dry particle diameter. Equation 12 describes the case where the spherical droplet formed completely covers the insoluble inclusion. The critical saturation ratio of such a droplet is lowered compared to a droplet formed on a particle without the insoluble part but the same amount of solute. Should the insoluble part be replaced by soluble material, the critical saturation ratio would be further lowered.

5. Cloud formation and precipitation

Clouds form in the atmosphere when the partial pressure of water vapour exceeds the saturation vapour pressure. These meteorological conditions can have several causes. The most common cloud formation appear in air that is moving upwards, e.g. lifting of air over mountains or fronts, or as a result of buoyant motions due to local heating. The upward motion induces expansion of the air due to decrease of pressure with altitude in the atmosphere. The expansion is almost adiabatic implying that the air cools as it rises. Lowered temperature causes a reduction of the water saturation vapour pressure. When we consider the effect on the humidity we find that the expansion in itself act to reduce and the induced cooling act to increase the saturation ratio. The net result for a rising and expanding air mass is that the saturation ratio increases with altitude. Should the air mass rise for a long time, eventually the saturation ratio will exceed unity and a cloud forms.

In cloud formation large amounts of water vapour condenses and form droplets. In principle these droplets could be formed by self nucleation, i.e. directly from the vapour phase without an nucleating condensed phase. This process requires a saturation ratio of approximately 10 (1000% relative humidity) to initiate droplet formation. These high saturation ratios are normally not

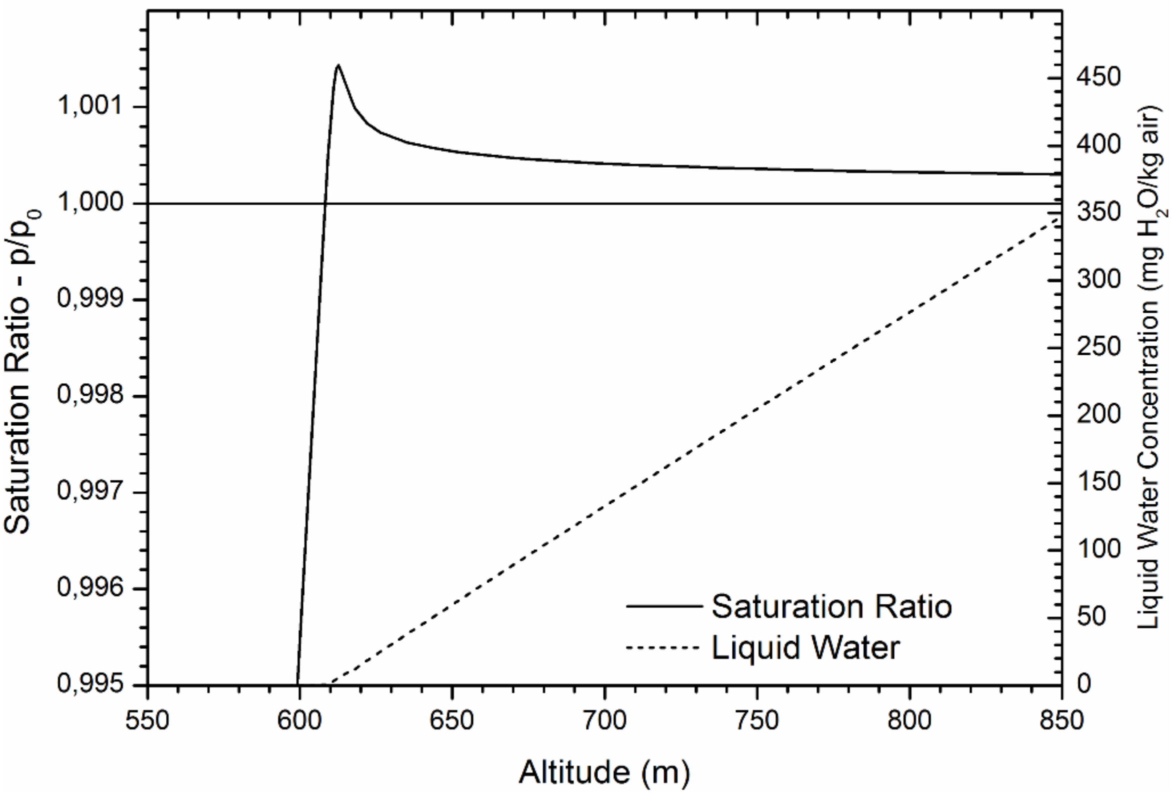
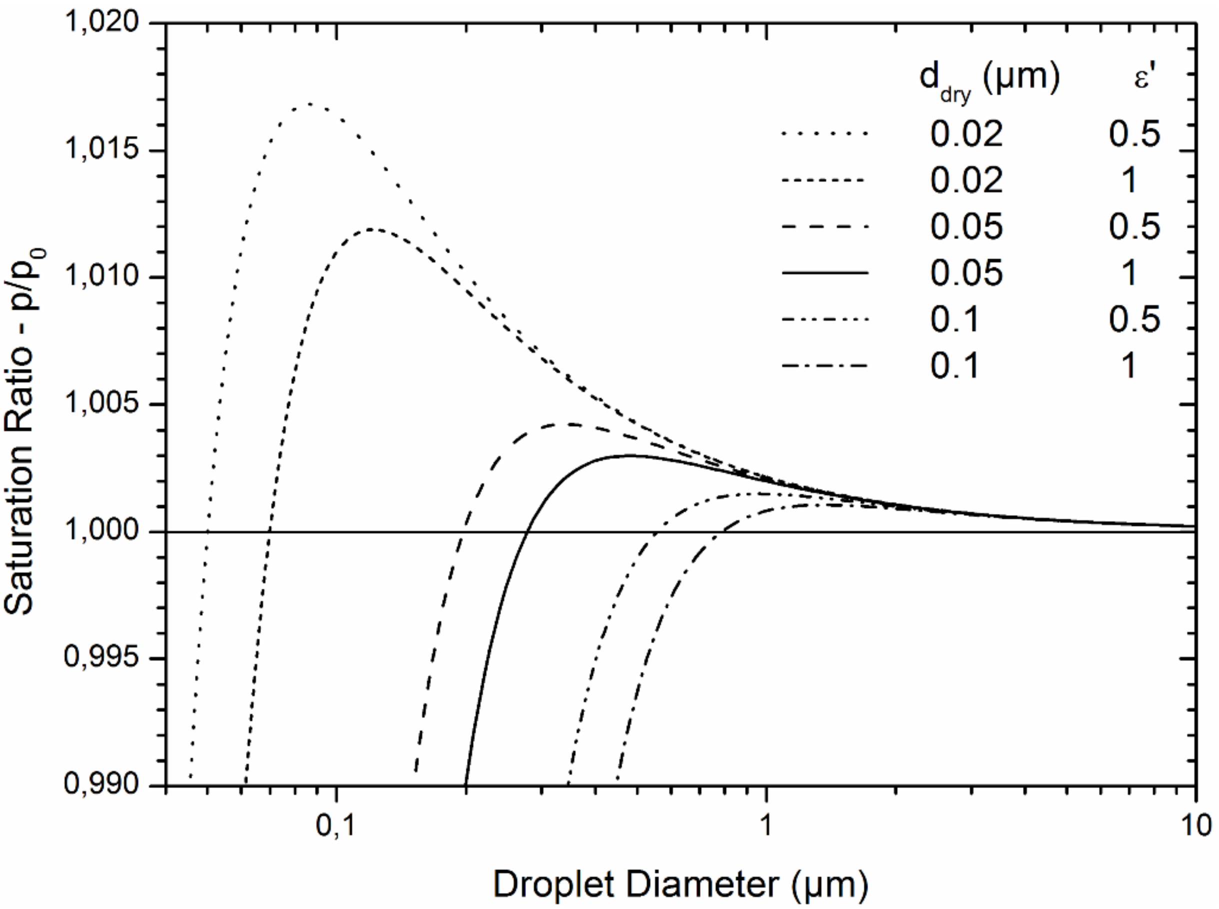


Figure 5. Saturation ratio and concentration of liquid water in an idealized cloud as a function of altitude. Data was obtained from a cloud model where the air mass was treated as a closed parcel and the updraft velocity was 0.1 m/s.

reached in the atmosphere because aerosol particles providing nucleated condensation are present everywhere in the atmosphere. This process consumes the water vapour in cloud formation long before self nucleation can start. As an example, the 0.05 µm NaCl particle in Figure 3 requires the saturation ratio 1.003 to form cloud droplets. We can thus conclude that ever-present aerosol particles act as condensation nuclei when clouds form in the atmosphere.

When the saturation ratio of a rising air mass exceeds unity it produces condensable water vapour, and continued rise produces more. Condensation on cloud droplets acts as a sink and consumes that water. The surface area available for condensation grows as the droplet size increases, implying that the sink becomes more efficient. As a result, the availability of condensable water reaches a maximum, after which it slowly decreases. Figure 5 shows an example of the vertical profile of the saturation of an idealized cloud formed in a rising air mass. The air mass reaches a maximum in saturation ratio slightly above the altitude where the cloud formation started. This level can be identified by the amount of condensed water in the cloud (Figure 5; the broken line). In this example the cloud formed at slightly above 600 m altitude. As the air mass continue to rise the cloud deepens and the amount of condensed water increases.

We will now turn our attention to the aerosol particles in the rising air mass. We know that the critical saturation ratio need to be exceeded for cloud droplet to form. This quantity depends on



Figur 6. The Köhler equation applied to six particles of different size and composition. The particles consists of pure NaCl (ε' = 1) or 50% (by volume) NaCl and 50% insoluble matter(ε' = 0.5).

particle properties. Figure 6 shows Köhler curves for six different particles calculated by equation 12. It is clear that the dependence on particle size is strong. The particle chemical composition is also important, which can be seen by comparing particles of the same size but with different composition in Figure 6. Let us now assume that these particles are involved in the cloud formation of Figure 5, where the maximum saturation ratio reached was 1.0014. We see that only one of the particles in Figure 6 has a critical saturation ratio lower than that. This implies that only one of the particle types, pure NaCl with 0.1 µm diameter, can act as cloud condensation nuclei and form large, activated cloud droplets in this cloud. The other particles remain in the cloud as small interstitial droplets that are bound to equilibrium size, e.g. at 800 m altitude in our example cloud the 0.02 µm diameter pure NaCl particle would be an interstitial droplet of approximately 0.075 µm diameter. The size-dependence can be studied in a diagram over the fraction of the particles of a given size that form cloud droplets (scavenging ratio) against the dry particle size (d_{dry}). In an ideal cloud containing particles of only a single composition this fraction rises from 0 to 1 at the diameter d'. If d is smaller than d' the fraction of cloud droplet is 0 and above d' it is 1. When the chemical composition varies among the particles, the region around d' where the scavenging ratio increases becomes broadened. In that case it is customary to set d' to the dry diameter where 50% of the particles form cloud droplets. When clouds are studied experimentally further broadening of the region around d’ where the scavenging ratio increases appears due to variable humidity history of probed air masses.

The particle size d’ where 50% of the particles act as condensation nuclei depends both on the properties of the aerosol particles and the cloud. Clouds with a high updraft velocity, like cumuliform clouds, produce larger amounts of condensable water per unit time than clouds with a lower updraft velocity. As a result the maximum saturation ratio becomes higher before condensation breaks the increase. In comparison with cloud of low updraft velocity, like stratiform clouds, cumuliform clouds reach higher maximum saturation ratio allowing more particles to form activated droplets. Increased updraft velocity of the cloud thus shifts d’ to smaller particle diameters. Should we instead change the aerosol properties for a given meteorological situation by increasing the number of particles that can act as cloud condensation nuclei, we would see that d’ shifts to larger diameter. This shift, however, is not that large that it compensates for the particles added, i.e. increased particle number concentration increases the number of cloud droplets. We see that human activities by increasing the particle number concentration can affect the microstructure of clouds, which is important for the radiative properties of clouds, and also for the formation of precipitation. A change of the cloud microstructure by human-induced change of the aerosol thus can affect the climate by the so called indirect effects of aerosol, which is described in the chapter on the climate.

Formation of cloud droplets is driven by diffusion of water vapour to the droplets. This diffusional growth is effective until the droplets reach approximately 20 – 30 µm diameter. For the formation of precipitation, i.e. to reach sufficient gravitational settling velocity, the droplets need to increase their diameter by approximately a factor of 100. The diffusional growth is not sufficiently fast and the availability of water vapour is not sufficient to form precipitation in this way. Instead precipitation is formed in a secondary step where cloud droplets or ice particles collide and merge to become larger. Precipitation formation starts by that some of the droplets or ice particles are somewhat larger than their neighbours. Their larger gravitational settling velocity increases the probability of collisions with their smaller and slower neighbours that are situated below. After collision and merging the new drop/ice particle has still larger settling velocity and further increased collision probability. The formation of precipitation need to overcome a threshold. Once started, the process accelerates. Most clouds in the atmosphere do not form precipitation. Their life cycle ends by a change of the meteorological conditions that reduces the saturation ratio to below unity, causing the cloud droplets to evaporate. Precipitation can more easily form in cold clouds (below freezing) than in warm ones as a result of that the former tends to form mixed phase clouds, thus containing mostly super-cooled droplet and a few ice particles. The saturation vapour pressure over ice is smaller than over liquid water (Figure 1). As a result ice particles grow faster, and can thus settle and collide with super-cooled droplets.

Short supplement: Climate effects of airborne particles

Some days it can be difficult to distinguish distant objects, whereas on other days details can easily be seen. This difference is caused by a variability in light scattering causing changes of the atmospheric visibility, which expresses our ability to distinguish distant objects. Gases are inefficient in scattering light. The varying visibility is mainly caused by airborne particles. Particles of approximately the same diameter or larger than the wavelength of light efficiently scatter light by Mie scattering. Often we observe reduced visibility during humid conditions. This is not caused by scattering by water vapour. Instead hygroscopic growth of aerosol particles is the cause of increased the scattering. The visibility can be reduced to a few meters in fogs where droplets in excess of 10 µm may form from sub-micrometer particles. Light scattering appears in all directions, implying that solar radiation can be scattered back to space by airborne particles without contributing energy to the earth by absorption. In this way airborne particles affect the planetary albedo. This effect on the radiation balance is called the direct effect of aerosol particles.

Clouds are important in the climate system being the cause a large fraction of the planetary albedo. Approximately 19% of the incoming solar radiation is scattered back to space by clouds. Human activities have increased the amount of particles in the atmosphere. These particles take part in cloud formation by serving as condensation sites for cloud droplet formation, resulting in increased number concentration of cloud droplets compared to an unpolluted cloud. This means that the cloud water will be distributed over a larger number of droplets, making the droplets smaller in size. However, the net effect of the increased number of droplets is increased surface area leading to increased scattering of light, thus increasing the cloud albedo. This is called the first indirect effect of airborne particles. There is also a second indirect effect. This effect is just like the first effect connected with the change of droplet number and size. Reduced droplet size lowers sedimentation velocities of the cloud droplets. Gravitational settling velocities are important for the formation of precipitation. As a result of the reduced droplet sizes, precipitation formation could be delayed or inhibited. This second indirect effect thus could cause increased cloudiness as a result of increased lifetime of the cloud, which means more time for such clouds to scatter solar radiation back to space.