

Replies to the comments of referee 1.

We are grateful to the referee for the constructive criticism, which helped to improve the clarity of the manuscript. Please find below the replies to the specific comments and an account of the modifications implemented.

1. *Section 2.1 would read better for me if the authors had first shown the solution for time-dependent vapour pressure (solutions given by Equations 10 & 11) followed by the simplified solutions pertinent to constant vapour pressures (Equations 4 & 7). This change is not necessary, but would be an improvement in my view.*

We changed Section 2.1 as suggested by the reviewer.

2. *Figure 9 compares model predictions with observations made on July 24 in Hyytiälä. A first order removal term ($\tau_{\text{loss}}=7$ hours) was included in the GDE to account the observed decrease in number concentrations observed at night (see Figure 7). Initial growth rates of 2.6 nm/hour at night and 12 nm/hour in the daytime were input parameters for the model. The authors argue that the model satisfactorily explains the nighttime and daytime observations of $CS(t)$ and $d_p(t)$, but not the large decrease in CS beginning at about 07:00, presumably due to breakup of the inversion after sunrise. While this might be an interesting heuristic result, I question its applicability to the atmosphere. More effort would be needed to demonstrate that all pertinent atmospheric processes have been properly taken into account. For example, does diameter increase linearly and monotonically with time during the daytime on all days at Hyytiälä? If not, it would appear that the authors picked a day with observations that could be explained, not necessarily because the model correctly describes what happened. I would be more comfortable with the paper if Figure 9 were deleted.*

Figure 9 is just an illustration of how the model works in the nearly ideal conditions, when the continuous aerosol growth by condensation can be clearly distinguished in the atmosphere. This holds for mostly sunny days and within one air mass. Generally, aerosol modes do not exhibit this well-pronounced growing dynamics, with the growth process being rather often interrupted either due to the changing air mass, precipitation or some other reason, which must be well understood and parameterized before they can be incorporated into the model. Within the time slots when aerosol mode grows due to condensation, our model can be successfully applied. We do not mean here to state that this model will work perfectly well for any day and any conditions. We would rather keep Figure 9 for illustrative purposes but we additionally stress the applicability of the model to idealized conditions and add this discussion in Lines 7-10, p. 10 and Lines 1-6, p.11.

3. *Figure 3 shows the relationship between CS calculated using the authors' simplified expression and the value of CS calculated using kinetic theory for a range of aerosol properties pertinent to the atmosphere. The default method for calculating CS for atmospheric data is by using the full FS expression, not the kinetic approach. The only rationale I can see for including this figure has to do with the relationship between the kinetic CS and the extinction coefficient mentioned in the conclusions. As that is not the focus of the paper, I would recommend either that Figure 3 be deleted (my preference) or that it be replaced with a figure that compares the CS_{corr} to CS_{FS} .*

Using Fig. 3 we wanted to demonstrate, besides the conclusion mentioned by the referee, for what

parameters it is relevant to use the corrected solution as compared to the kinetic regime. In the kinetic regime, as could be seen from our analysis and elsewhere, all the formulas are very simple. It might well be that for some organic vapours, characterized by relatively large molecular weights as compared to sulfuric acid, and, consequently, large mean free paths, the kinetic regime formulas might work reasonably well. Thus, it seems more relevant to compare here the kinetic regime and the correction. That is one of the reasons we would like to keep Fig. 3: in order to give the reader a possibility to estimate the parameters and find out what regime can be used when the chemical formula of the vapour is known (or its molecular weight can be estimated approximately).

Moreover, in spite of the paragraph in the conclusion regarding this figure being indeed not really in the focus of this paper (but still related to this problem), it is still an important note. The question whether CS is a suitable parameter to characterize scatter of light on aerosols is quite important while performing general multidisciplinary analyses on atmosphere-biosphere interaction. The latter usually report the results combining several problems on different topics in one and it would be cumbersome to give all the details in one paper. That is why we would prefer to keep this figure in this paper and to refer to this paper when needed.

Minor points:

1. *I recommend that in the first sentence following equation (1) the authors explicitly define $n_d = n_d(t, d_p) = dN/dd_p$. Other forms of the distribution function are often used (see, for example, Figures 4, 5, 7 of this paper).*

The point is changed as recommended.

2. *Sentence following Equation (5): should be "down to $Kn \approx 0.5$ ", not "up to $Kn \approx 0.5$ ". The approximation is valid in the high Kn (low $1/Kn$) range.*

The point is changed as recommended.

3. *Line 27, page 7: "The correction results in a 5.5% increase in CS, in accordance with Fig. 1." I suggest rewording this sentence. Figure 1 shows that for $Kn > 0.5$, the approximate expression for CS exceeds the full expression by an amount ranging from 0 to 8%. So the direction is consistent and the observed 5.5% discrepancy is consistent. That's all we know for certain.*

We agree with the comment. This sentence now reads: «The correction results in a 5.5% increase in CS, which is consistent with the increase in the mass flux from 0 to 8% as compared to the full FS formula (Fig. 1).»

4. *p. 8, line 1: "reflects the error due to approximation." The paper addresses several approximations. The text should be clarified to specify which approximation is responsible for the large differences in this case.*

We agree with the comment. This sentence is now rephrased to: «reflects the error due to approximating the measured particle number size distribution with a lognormal distribution».

5. *p. 8, line 5: "(see Fig. 4, right panel, at 00:30 to 02:00, when the aerosol mode characterized by low number concentration but large particle diameters sporadically appears)." I have gone back to look for this several times in Figure 4. It is not apparent to me.*

We added the following sentence: «In the left panel this mode is almost not visible due to the non-logarithmic scale of the particle number distribution contour map.»

6. p. 9, line 2: *"feature makes is difficult" should be " feature makes it difficult"*

We changed this in the text.

7. *Figures 4, 5 & 7 compare values of CS calculated from "measurements" (green lines) with those calculated from "theory [i.e., Eq. 16]" (red lines). For clarity the authors should revise this terminology for the following reasons: (1) both of these calculations require the use of measurements, (2) the results calculated directly from measurements (green) are more theoretically correct than those calculated from the simplified model (red). I believe this is shorthand language the authors used to communicate among themselves, but which is not helpful for the reader.*

We agree with the comment. Instead of «measurements» we use now «definition», and instead of «theory» we use «simplified model».

We thank again the referee for the useful suggestions. We hope that you will find that the present manuscript addresses all the comments raised.

Replies to the comments of referee 2.

We are grateful to the referee for the constructive criticism, which helped to improve the clarity of the manuscript. Please find below the replies to the specific comments and an account of the modifications implemented.

On page 3 just before equ. 3 it is stated that the "...FS coefficient ... connects the mass flux towards a molecule in the kinetic regime...". I believe it should be the "mass flux towards a particle".

We agree with the comment and changed this as recommended.

On page 6, lines 20/21, the authors mention that equ. (15) and (16) can be used in quasi-steady conditions together with distribution parameters changing slowly in time. To get some idea it would be helpful here if corresponding time scales would be mentioned. There is some indication given in a later section but should also be discussed here.

We agree with the comment and add the sentence regarding time scales: «...change slowly in time (as discussed later, the typical time scale of the system relaxation to the equilibrium is expected to be not more than 20 min, hence, the time scale on the order of hours or diurnal scale can be used to define the slow change in this context).»

Some clarification is needed in the discussion of Figure 6 on page 7, lines 26-31. Fig. 6 displays CS for several days but then it is referred to "note that $Kn \sim 5.2$ on March 27th and $Kn \sim 2.5$ on June 1st" (line 30). I don't see how this connects to Fig. 6, at least it is not visible to me.

We agree with the comment. We changed this sentence to the following: «Note that the Knudsen numbers corresponding to the geometric mean diameters of the modes used for calculations are $Kn > 2.5$, and the largest particles taken into account here ($d_p = 500$ nm) have $Kn \sim 0.5$, hence, the correction should perform well, as follows from Fig. 1.»

Figure 3: Please expand figure caption, some more explanation is needed here. Also, no reference is made to the color code (condensation sink ratio).

We agree with the comment. The caption is now expanded to read: «The ratio $CS_{cor,0} = CS_{kin,0}$ as a function of σ_0 (characterizing the width of the particle number-size distribution) and the Knudsen number Kn_0 (corresponding to the mean geometric diameter of the mode).» We added the reference to the color code.

Figure 8: Again, more elaborate figure caption would be desirable. What are the lines representing? BTW: the inset is hard to read.

We changed figure 8 as recommended. The caption is changed to read: «A diagram showing the condensation sink, CS, as a function of the geometric mean diameter of the aerosol mode, d_{p0} , and the particle number concentration, N_0 , for $\sigma_0 = 1.5$.»

Figure 9: line types dashed and dotted are hard to distinguish.

We agree with the comment and changed the line types in figure 9.

Editorial comments:

Page 2, lines 18/19: "...affects little the particle growth" (delete "to")

We changed this point as recommended.

Page 7, line 27: "...showing that the mass..." (delete one "the")

This sentence has been changed due to the comment of the other reviewer.

Page 9, line 6: "...time scales..."

We changed this point as recommended.

We thank again the referee for the useful suggestions. We hope that you will find that the present manuscript addresses all the comments raised.

A list of relevant changes:

1. Section 2.1 is re-ordered in accordance with the suggestions of Referee 1.
2. In the figures, we increased font size in labels and line widths.
3. We changed the captions to Figures 3 and 8.

Minor changes are marked in blue in the text.

A simple model for the time evolution of the condensation sink in the atmosphere for intermediate Knudsen numbers

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Abstract. Transformation of the mass flux towards the particle from the kinetic regime to the continuum regime is often described by the Fuchs-Sutugin coefficient. Kinetic regime can be obtained as a limiting case when only one term of the expansion of the Fuchs-Sutugin coefficient at small $1/Kn$ is considered. Here we take into account the two first terms, and get the mass flux which agrees well with the full mass flux down to $Kn \sim 0.5$. This procedure allows to obtain an analytical solution of the condensation equation valid for the range of intermediate Knudsen numbers. The expansion is further applied to calculate analytically the condensation sink. The formula for the condensation sink is tested against field observations. The relative contribution of different aerosol modes to the condensation sink is discussed. Furthermore, we present a simple model describing a coupled dynamics of the condensing vapour and the condensation sink. The model gives reasonable predictions of the condensation sink dynamics during the periods of the aerosol modes growth by condensation in the atmosphere.

10 1 Introduction

Condensation sink (CS) is an important parameter for aerosol dynamics quantifying the rate of the vapour condensation on the existing aerosol population. The inverse of CS has a clear physical meaning being the characteristic time scale for vapours to condense onto the surface of existing aerosol. Due to the similarity between the processes of vapour condensation on aerosol particles and coagulation of the smallest particles (monomers, dimers, clusters) with the larger particles from Aitken and accommodation modes, CS proves useful for the quantification of a coagulation sink (Lehtinen et al., 2007). Then a competition between the process of small clusters coagulating with the larger aerosol particles, represented by CS, and the process of the clusters growth by condensation, represented by particle growth rate, defines the probability of clusters survival and a new particle formation event (Kulmala et al., 2017). A detailed comparative analysis of physical processes in the atmosphere based on the characteristic timescales was performed by Kerminen et al. (2004).

20 The average condensation sink on new particle formation event days is generally lower as compared with nonevent days (Kulmala et al., 2001; Birmili et al., 2003; Hyvönen et al., 2005; Wu et al., 2007; Asmi et al., 2011; Pikridas et al., 2012; Young et al., 2013; Kanawade et al., 2014; Qi et al., 2015; Vana et al., 2016). Indeed, a large condensation sink means that a relatively large surface of aerosol is available for condensation and coagulation with clusters serving as precursors of newly forming particles. However, at highly polluted places such as megacities, the new particle formation events happen even for

large CS (Wu et al., 2007; Xiao et al., 2015). The dynamics of CS is tightly connected with different atmospheric processes, including the effects of atmospheric chemistry, meteorology and solar radiation. A simple model allowing to describe the dynamics of a condensation sink in the atmosphere could thus be helpful for understanding of new particle burst and cut-off processes. Here we develop a basis for such a model.

5 For describing aerosol dynamics, we choose a modal approach. This approach treats the whole aerosol population as a sum of modes, and the equations for the first-order moments of the particle size distribution are obtained based of the aerosol general dynamics equation. The number concentration, geometric mean diameter and standard deviation of each mode can be calculated from the moments (Whitby and McMurry, 1997). Assuming a particular type of the particle size distribution (usually lognormal) makes it possible to get a closed system of equations for the moments. This method is not too expensive
10 computationally, and is at the same time rather accurate (Whitby and McMurry, 1997). A ‘pseudomodal’ approach has been used to develop fast and efficient aerosol microphysics modules for large-scale atmospheric modelling purposes (Vignati et al., 2004; Stier et al., 2005; Mann et al., 2010; Pringle et al., 2010; Aquila et al., 2011; Zhang et al., 2012; Liu et al., 2016).

Instead of using the full general dynamics equation, we focus here on one physical process - aerosol growth by condensation - because of its importance for atmospheric aerosol. The model developed here is tested against atmospheric observations
15 from a remote site Hyytiälä (Finland), representing semi-clean boreal forests in the Northern hemisphere. Typically one can identify two or three modes with the characteristic diameters less than 200 nm at this site (dal Maso et al., 2008). Both day and night aerosol population behaviour clearly demonstrate patterns typical for condensational growth. On the contrary, another important physical process, coagulation, while a potentially important sink for growing clusters and nanoparticles, affects little the particle growth rate unless the number concentration of the growing particle populations is very high (Kerminen et al.,
20 2004).

Aerosol growth by condensation has been extensively investigated theoretically (e.g. Kulmala, 1993; Vesala et al., 1997; Park and Lee, 2000). Besides the simple formulations for the rates of growth involving different physical phenomena at different scales (e.g., Barrett and Clement, 1988), there are models describing the coupled dynamics of the vapour concentration and aerosol distribution applied to the processes in the atmosphere (Clement et al., 2001) and aerosol chambers (Stock, 1987;
25 Barrett et al., 1992; Wu and Biswas, 1998). Most of these models, however, are still quite complicated.

The novelty of the present work is that we obtain analytical formulas for the condensation sink and its time evolution in the range of intermediate Knudsen numbers typical for the atmospheric applications. Two regular approaches which can be found in the literature involve either extensive calculations starting with Boltzmann equations (e.g., Kosuge et al., 2010) or employ matching functions (Barrett and Clement, 1988), giving the correct expressions in the molecular and continuum limits
30 and ‘something in between’ in the transitional regime (unless the full Fuchs-Sutugin coefficient is applied, making further analysis possible only by numerical methods). Park and Lee (2000) showed that the latter method (harmonic mean) results in a mass flux quite similar to the one obtained with the full Fuchs-Sutugin coefficient (Fuchs and Sutugin, 1971). They obtained an analytical solution of the condensation equation valid for the whole range of diameters. However, this solution is quite complicated and can not be integrated to get an analytical expression for CS. Here we proceed using the first two terms of the
35 expansion of the Fuchs-Sutugin coefficient in terms of $1/Kn$ for small $1/Kn$. As can be seen later, this makes it possible to find

analytical formulas for the evolution of the particle size distribution and CS for the intermediate range of Knudsen numbers, while remaining close to those calculated using the original Fuchs-Sutugin coefficient. The expansion is in agreement with the full formula for particle diameters up to ~ 450 nm, i.e. for Aitken mode and almost the whole range of sizes typical for the accumulation mode, not only in remote places like a typical boreal forest (Hyytiälä, Finland), but also in megacities, such as

5 Beijing in China (e.g. Liu et al., 2017).

Analysis of the time scales typical for the dynamics of CS and vapour concentration in the atmosphere, allows to use a quasi-stationary approach for the vapour concentration and develop a simple model describing the coupled dynamics of the CS and the condensing vapour in the atmosphere during the periods of aerosol growth by condensation inherent in the atmosphere.

2 A theoretical model

10 2.1 The kinetic regime vs the intermediate regime

The equation describing the growth of the aerosol population by condensation is

$$\frac{\partial n_d}{\partial t} = -\frac{\partial(I_d n_d)}{\partial d_p}. \quad (1)$$

Here $n_d = n_d(t, d_p) = dN/dd_p$ is the particle number distribution (N is the particle number concentration), t is time, d_p is the diameter of a particle and I_d is the growth rate. The growth rate can be written as follows (Seinfeld and Pandis, 2016):

$$15 \quad I_d(d_p, t) = \frac{\alpha M_A v_c (p(t) - p_{eq})}{2\rho_p RT} \beta_1, \quad (2)$$

where α is the mass accommodation coefficient of the condensing vapour, M_A is its molar mass, v_c is the mean speed of the vapour molecules, $p(t) - p_{eq}$ is the difference between the vapor pressure far from the particle and the equilibrium vapour pressure (generally time-dependent), ρ_p is the particle density, R is the universal gas constant and T is temperature. The Fuchs-Sutugin (FS) coefficient, β , is generally used to correct the mass flux towards a particle in the continuum regime in order to

20 get a smooth transition of the mass flux from the continuum to the kinetic regime. For the purposes of the present study, it is convenient to introduce a modified Fuchs-Sutugin coefficient, β_1 , which corrects the mass flux in the kinetic regime and provides a smooth transition of the mass flux in the kinetic regime to that in the continuum regime:

$$\beta_1 = \frac{4Kn(1 + Kn)}{3\alpha(1 + 0.377Kn + 1.33Kn(1 + Kn)/\alpha)}. \quad (3)$$

Here the Knudsen number is $Kn = \frac{2\lambda}{d_p}$ where λ is the free mean path of condensing molecules. The solutions of the condensation equation have been extensively investigated (e.g., Clement, 1978) and the method of characteristics has proven to be

25 a useful tool.

In the kinetic regime $Kn \gg 1$, $\beta_1 = 1$, thus I_d does not depend on the particle diameter. The solution of equation (1) in the kinetic regime obtained with the method of characteristics is

$$n_d(d_p, t) = n_{d0}(d_p - \int_0^t I_{d,\text{kin}}(t') dt'), \quad (4)$$

where $I_{d,\text{kin}}(t') = \frac{\alpha M_A v_c (p(t') - p_{\text{eq}})}{2\rho_p RT}$ and n_{d0} is the distribution of particles at initial time $t = 0$. If the mode is initially lognormal with the geometric mean diameter of d_{p0} , this solution prescribes the growth of the characteristic diameter of the distribution linearly in time without any change in the shape of the distribution.

In the next order in $(1/Kn)$, one obtains from (3):

$$\beta_1 = 1 - \frac{0.377\alpha}{1.33Kn}. \quad (5)$$

This function is shown in Fig. 1 together with the full FS coefficient and the kinetic regime approximation, $\beta_1 = 1$. Formula (5) shows a good correspondence with the full formula (3) down to $Kn \approx 0.5$, with the overestimation of the mass flux towards the particles not more than 8%. As (5) is obtained from the kinetic regime formula by account of the term of the next order of smallness, we refer to it further as ‘correction’. In this case the growth rate is not constant but depends on the particle diameter:

$$I_{d,\text{corr}}(d_p, t) = I_{d,\text{kin}}(t) \left(1 - \frac{0.377\alpha d_p}{2.66\lambda}\right), \quad (6)$$

which means that the larger particles grow slower than the smaller particles. This difference leads to the narrowing of the initial distribution with time.

One can introduce a limiting diameter as $1/d_{\text{lim}} = \frac{0.377\alpha}{2.66\lambda}$, which corresponds to a zero mass flux towards the particle. Note that for the accommodation coefficient $\alpha = 1$ the limiting diameter is on the order 7λ and $Kn \approx 0.28$, which is beyond the range of $Kn \gtrsim 0.5$, where the correction can be applied. Thus, the diameters corresponding to a non-physical zero mass flux will not be considered in the framework of the present model.

The solution of the condensation equation obtained with the method of characteristics for intermediate Knudsen numbers is

$$n_d(d_p, t) = \exp\left(\frac{\int_0^t I_{d,\text{kin}}(t') dt'}{d_{\text{lim}}}\right) n_{0d} \left(d_{\text{lim}} + (d_p - d_{\text{lim}}) \exp\left(\frac{\int_0^t I_{d,\text{kin}}(t') dt'}{d_{\text{lim}}}\right)\right). \quad (7)$$

The solutions of the condensation equation (4) and (7) can be simplified for the constant pressure difference. The growth rate can be written as follows

$$I_{d,\text{kin}} = \frac{\alpha M_A v_c (p - p_{\text{eq}})}{2\rho_p RT} \beta_1 \quad (8)$$

and the solution in the kinetic regime is:

$$n_d(d_p, t) = n_{d0}(d_p - I_{d,\text{kin}}t), \quad (9)$$

while applying the correction leads to the solution:

$$n_d(d_p, t) = \exp\left(\frac{I_{d,\text{kin}}t}{d_{\text{lim}}}\right) n_{0d}\left(d_{\text{lim}} + (d_p - d_{\text{lim}}) \exp\left(\frac{I_{d,\text{kin}}t}{d_{\text{lim}}}\right)\right). \quad (10)$$

- 5 We next compare the kinetic limit solution and the ‘corrected’ solution for the constant growth rate of the particles $I_{d,\text{kin}} = 7$ nm/h, typical for a continental boundary layer during summer time (Yli-Juuti et al., 2011), taking the initial lognormal particle number distribution:

$$n(d_p) = \frac{N_0}{\sqrt{2\pi}d_p \ln \sigma_0} \exp\left(-\frac{\ln^2(d_p/d_{p0})}{2\ln^2 \sigma_0}\right). \quad (11)$$

- The mean free path for the molecules of sulphuric acid, which can be considered as a typical low-volatile condensing vapour, is $\lambda \approx 120$ nm. Taking the accommodation coefficient as $\alpha = 1$, the geometric mean diameter at $t = 0$ s as $d_{p0} = 20$ nm, particle number concentration as $N_0 = 2000 \text{ cm}^{-3}$ and $\sigma_0 = 1.5$, we obtain the solutions in Fig. 2. As was mentioned earlier, the correction leads to a decrease in the growth rate as compared to the kinetic regime, and the distribution shape changes in time if the correction is applied. Note that the solution obtained numerically employing the full FS coefficient (shown with symbols in Fig. 2) is in close agreement with that given by eq (10).

15 2.2 Condensation sink

In this Subsection we apply the correction to obtain analytical formulas for the condensation sink. The CS reflects the ability of vapours to condense on the aerosol particles and can be calculated from the formula (e.g. Kulmala et al., 2001):

$$\text{CS} = 2\pi D_v \int_0^{d_{p,\text{max}}} d_p \beta n(d_p) dd_p, \quad (12)$$

where D_v is the diffusion coefficient of the condensing vapour and $\beta = 3\alpha\beta_1/4Kn$.

- 20 Assuming that dynamics of an aerosol mode is described by solution (9) and taking the initial lognormal distribution (11), in the kinetic regime the integration of (12) yields:

$$\text{CS}_{\text{kin}} = \frac{2\pi D_v d_{p0}^2 N_0}{\lambda} \frac{\alpha}{2.66} \left(\exp(2\ln^2 \sigma_0) + 2 \left(\frac{I_{d,\text{kin}}t}{d_{p0}}\right) \exp(0.5\ln^2 \sigma_0) + \left(\frac{I_{d,\text{kin}}t}{d_{p0}}\right)^2 \right). \quad (13)$$

It can be seen that the CS grows in time as t^2 , which is not surprising given that the CS is proportional to the surface area available for condensation (i.e., the total surface area of the aerosol population), and the diameter of each particle grows linearly in time.

Accounting for the correction valid for intermediate Knudsen numbers gives $\beta = \frac{\alpha d_p}{2.66\lambda} - \frac{0.377}{4} \left(\frac{\alpha}{1.33}\right)^2 \left(\frac{d_p}{\lambda}\right)^2$. Thus, taking the initial lognormal distribution (11) and integrating formula (12) in view of solution (10), we obtain the CS evolution in time:

$$\begin{aligned} \text{CS}_{\text{cor}} = & \frac{2\pi D_v d_{p0}^2 N_0}{\lambda} \frac{\alpha}{2.66 \exp\left(\frac{2I_{d,\text{kin}}t}{d_{\text{lim}}}\right)} \left(e^{2\ln^2 \sigma_0} - 2\frac{d_{\text{lim}}}{d_{p0}} f(t) e^{0.5\ln^2 \sigma_0} + \left(\frac{d_{\text{lim}}}{d_{p0}}\right)^2 f^2(t) - \right. \\ & - \frac{0.377\alpha}{1.33Kn_0 \exp\left(\frac{I_{d,\text{kin}}t}{d_{\text{lim}}}\right)} \left(e^{4.5\ln^2 \sigma_0} - 3\left(\frac{d_{\text{lim}}}{d_{p0}}\right) f(t) e^{2\ln^2 \sigma_0} + \right. \\ & \left. \left. + 3\left(\frac{d_{\text{lim}}}{d_{p0}}\right)^2 f^2(t) e^{0.5\ln^2 \sigma_0} - \left(\frac{d_{\text{lim}}}{d_{p0}}\right)^3 f^3(t) \right) \right), \end{aligned} \quad (14)$$

where $f(t) = 1 - \exp\left(-\frac{\int_0^t I_{d,\text{kin}}(t') dt'}{d_{\text{lim}}}\right)$, $Kn_0 = 2\lambda/d_{p0}$. Note that one can easily obtain the formulas for the CS similar to (13) and (14) if the vapour pressure varies in time, using the substitution $I_{d,\text{kin}}t \rightarrow \int_0^t I_{d,\text{kin}}(t') dt'$.

If the parameters of the aerosol population do not depend on time, CS in the kinetic regime can be calculated as follows:

$$\text{CS}_{\text{kin},0} = \frac{2\pi D_v d_{p0}^2 N_0}{\lambda} \frac{\alpha}{2.66} \exp(2\ln^2 \sigma_0), \quad (15)$$

The analogous formula defining CS for the intermediate Knudsen numbers is

$$\begin{aligned} \text{CS}_{\text{cor},0} = & \frac{2\pi D_v d_{p0}^2 N_0}{\lambda} \frac{\alpha}{2.66} \exp(2\ln^2 \sigma_0) \left(1 - \frac{0.377\alpha}{1.33Kn_0} \exp(2.5\ln^2 \sigma_0) \right) = \\ & = \text{CS}_{\text{kin},0} \left(1 - \frac{0.377\alpha}{1.33Kn_0} \exp(2.5\ln^2 \sigma_0) \right). \end{aligned} \quad (16)$$

Note the term in brackets, similar to the limiting diameter in the formula for the growth rate (6), but including also the width of the distribution. Clearly, the CS should not be zero and the largest σ_0 particles in the distribution, making a significant contribution to the CS, should have Knudsen numbers larger than 0.5 not to introduce errors.

In order to demonstrate the influence of correction (5) on CS, we compare the sinks calculated using formulas (15) and (16). Here we investigate the difference between the kinetic regime and correction, while the difference between the correction and the full FS formula for CS will be addressed in the next Section. The ratio of CSs, $\text{CS}_{\text{cor},0}/\text{CS}_{\text{kin},0}$, as a function of Knudsen number (based on the geometric mean diameter of the distribution) and σ_0 is displayed in Fig. 3. The difference between β_0 and β_1 in Fig. 1 is no more than 40%. Given that the coefficient β appears in the integral (12), the range of parameters

corresponding to $CS_{\text{cor},0}/CS_{\text{kin},0} \leq 0.6$ should be disregarded. This range, as can be seen from Fig. 3, includes small Knudsen numbers and large σ_0 , i.e. wide distributions with large geometric mean diameters. Obviously, for this range of parameters a part of the distribution is beyond the limits of the correction applicability.

Formulas (15) and (16) can be used in quasi-stationary conditions, when the aerosol distribution at every time moment can be approximated by the lognormal one and parameters defining the distribution, d_{p0} , σ_0 and N_0 , change slowly in time (as discussed later, the typical time scale of the system relaxation to the equilibrium is expected to be not more than 20 min, hence, the time scale on the order of hours or diurnal scale can be used to define slow change in this context). Moreover, CS for several modes can be calculated as a sum of the CSs for each of the modes. Note also that the formulas obtained in this section are valid for the majority of atmospheric conditions, unless the coagulation process is important and the coagulation term has to be added in the GDE (for example, in highly polluted areas). We use formulas (16) for testing the theory against the atmospheric measurements in the next section.

3 Comparison with atmospheric observations. Quasi-stationary conditions and a weakly growing mode.

We first evaluate formula (16) using experimental data for quasi-stationary conditions or weakly growing modes (see examples in Figs 4, 5). The experimental data used for analysis in this Section are from the University of Helsinki SMEAR II station (Hari and Kulmala, 2005). Fig. 4 shows the particle size distribution in Hyytiälä on March 27th, 2014. From 00:30 to 07:00, local time, the aerosol mode remains almost unchanged, with the parameters weakly fluctuating around their mean values, and we refer to these conditions as quasi-stationary. During the daytime, there were growing Aitken and accumulation modes, but we use the same formula (16) and account only for the accumulation mode. As a second example, we consider the particle size distribution in Hyytiälä on 01.06.08 (Fig. 5), again using formula (16) for one mode with larger particles.

To calculate the CS from eq (16), we fitted the measured particle size distribution $dN/d(\log_{10} d_p)$ at different times by the lognormal distribution,

$$\frac{dN}{d(\log_{10} d_p)} = \frac{N_0 \ln(10)}{\sqrt{2\pi} \ln \sigma_0} \exp\left(-\frac{\ln^2(d_p/d_{p0})}{2 \ln^2 \sigma_0}\right). \quad (17)$$

The parameters N_0 , σ_0 and d_{p0} were obtained using the nonlinear least-squares Marquardt-Levenberg algorithm implemented in Gnuplot (Williams et al., 2013).

The parameters of the particle size distributions on March 27th, 2014 and June 1st, 2008 are summarized in Table 1, and the examples of experimental data fitting by the function (17) are shown in the lower panel of Fig. 5.

The performance of the analytical formula (16) is demonstrated in Figs 4, 5, right panels. The deviation of the theoretically calculated CS from the one obtained from experimental data using full FS coefficient is larger when the tails corresponding to the larger particles are not captured, while smaller particles seem not to be important. These examples illustrate the importance of the largest particles contribution to the CS (Lehtinen et al., 2003).

We next aim to separate the errors introduced by the insufficiently good approximation of the particle number distribution and usage of (5) instead of the full FS formula. We consider the fits of experimental data by the lognormal distribution and

calculate the CSs: 1) using full FS coefficient, and 2) using approximations of FS coefficient in the kinetic and intermediate regimes. Fig. 6 shows CS for several days in Hyytiälä in spring and summer, calculated from formulas (15) and (16), versus the full CS, eq (12). The correction results in a 5.5% increase in CS, which is consistent with the increase in the mass flux from 0 to 8% as compared to the full FS formula (Fig. 1). At the same time, the kinetic regime formula leads to overestimates of up to 20-25% for larger values of CS. Note that the Knudsen numbers corresponding to the geometric mean diameters of the modes used for calculations are $Kn \geq 2.5$, and the largest particles taken into account here ($d_p = 500$ nm) have $Kn \approx 0.5$, hence, the correction should perform well, as follows from Fig. 1. Thus, we can conclude that the large difference between the theoretical and experimental CSs in Fig. 5 reflects the error due to approximating the measured particle number size distribution with a lognormal distribution.

Overall, when considering the size range < 500 nm, the CS calculated using the formula (16) underestimates the CS obtained directly from measurements on average by 10-15%, and even more when the particle number distribution corresponding to larger particles is not captured well (see Fig 4, right panel, at 00.30-02:00 when the aerosol mode characterized by low number concentration but large particle diameters sporadically appears. In the left panel this mode is almost not visible due to the non-logarithmic scale of the particle number distribution contour map). This is to emphasize that not always one mode is enough for the good representation of the CS, even when the mode seems to be clearly prevailing. Fig. 7 shows an example of such a day. The orange curve in Fig. 7 corresponds to the theoretical calculations with one mode having the largest number concentration, while the blue curve shows the CS calculated for the two modes (as a sum, based on the approximation of experimental data by two modes). Clearly, both modes have to be accounted in this case.

One can get an estimate on the contribution of different modes to CS using a parameter map in Fig. 8. This map shows CS calculated using formula (16) as a function of d_{p0} and N_0 for the fixed $\sigma_0 = 1.5$. Typical Hyytiälä parameters in spring and summer give CS values between 0.001 and 0.01 s^{-1} (Dada et al., 2017). Modes with relatively small number concentrations ($\sim 100 \text{ cm}^{-3}$) and large characteristic diameters are likely to contribute significantly to CS, and the discrepancy between the measured and theoretically calculated CS for one mode is usually due to not accounting for these large particles. At the same time, the concentrations of smallest particles ($\sim 2 - 3$ nm) in the cluster mode can be very high in the atmosphere during nucleation events, (up to $10\,000 \text{ cm}^{-3}$ in Hyytiälä), yet they contribute little to the CS until they grow to sufficiently large diameters (~ 20 nm). Even when the particle number concentrations in the cluster mode (2-3 nm) are as high as 10^5 cm^{-3} (Kontkanen et al., 2017), their contribution to the typical atmospheric condensation sinks is negligibly small. Note that the maximum characteristic diameter shown here is 350 nm. This is due to the fact that our formula is valid for the particle diameters up to 450-500 nm. Thus, we are not able to draw the conclusions about the contributions of the supermicron modes based on the present theory.

4 Dynamics of aerosol mode growing by condensation

A coupled model of aerosol mode growing by condensation includes 2 equations:

1) equation of condensation:

$$\frac{\partial n_d}{\partial t} = -\frac{\partial(I_d n_d)}{\partial d_p}, \quad (18)$$

2) equation describing the time evolution of the vapour concentration:

$$\frac{dC}{dt} = Q(t) - CS \times (C - C_{\text{eq}}), \quad (19)$$

5 where C is the vapour concentration, C_{eq} is the equilibrium vapour concentration and $Q(t)$ is the rate of vapour production. The system is coupled in a sense that the equation for the particle number distribution includes the dependence on the vapour concentration through the growth rate as $I_d \sim (p - p_{\text{eq}}) \sim (C - C_{\text{eq}})$ and at the same time the equation for the vapour concentration contains the term with CS , proportional to the integral of the number particle distribution. This feature makes it difficult to solve eqs (18) and (19) simultaneously. However, as we showed before (Sec. 2), for the intermediate Knudsen numbers
 10 $Kn \leq 0.5$ equation (18) can be integrated and, assuming a lognormal distribution, an analytical formula for the evolution of the CS can be readily obtained, even for the case of the vapour concentration changing in time.

At the same time, both CS and vapour concentration typically change over [time scales considerably longer than 5-17 min](#).

In what follows we consider for simplicity non-volatile vapours with $C'_{\text{eq}} = 0$. We proceed to show that eq (19) can also be significantly simplified for the typical atmospheric values of CS . The lowest values of CS in Hyytiälä in spring and summer are
 15 around 0.001 s^{-1} (Dada et al., 2017), corresponding to the time scale $\tau \sim 1/CS = 17 \text{ min}$. Generally, however, values of CS are higher and the corresponding time scales are smaller than 17 min ($\sim 5 \text{ min}$ for Hyytiälä). At the same time, both CS and vapour concentration typically change over times scales considerably longer than 5-17 min (e.g., Petäjä et al., 2009; Kontkanen et al., 2016). This means that the solution of equation (19) relaxes fast (with a time scale $1/CS$) to the quasi-stationary regime, with

$$20 \quad C(t) = Q(t)/CS(t). \quad (20)$$

This formula is often used to get the proxies for vapour concentrations (Petäjä et al., 2009), and a similar expression, with corrections due to the time evolution of $Q(t)$ and $CS(t)$, has been used by Clement et al. (2001) for the analysis of the particle formation processes in Hyytiälä.

Thus, the system of two differential equations can be reduced to a relatively simple system of two algebraic equations:

25 1) eq (14) describing the CS evolution for the intermediate Knudsen numbers (or eq (13) in the kinetic regime) using the substitution $I_{d,\text{kin}} t \rightarrow \int_0^t I_{d,\text{kin}}(t') dt'$,

2) eq (20) for the vapour concentration,

and self-consistent dynamics of the system can be obtained from the simple iterations of this system.

Practically, one can start with some initial growth rate, and then calculate the increase in CS using eq (14) during the short
 30 period of time, $\delta t \sim 6 - 10 \text{ min}$. Then the vapour concentration can be found using eq (20) for the new CS , $CS(t_0 + \delta t) =$

$CS_0 + \delta CS$, at the next time step. The growth rate at this time step can be found as $I(t_0 + \delta t) = \frac{I(t_0)}{CS(t_0 + \delta t)/CS_0}$. If the production rate of condensing vapour is a function of time, then $I(t_0 + \delta t) = \frac{I(t_0)Q(t_0 + \delta t)/Q_0}{CS(t_0 + \delta t)/CS_0}$. Further, the increase in the CS is calculated from (14) with the new growth rate, and so on.

This procedure can be readily extended for the two aerosol modes if the CS is taken as the sum of the CSs calculated for each of the modes. The results of the model calculations with two modes is shown in Fig. 9 for one day in Hyytiälä (July 24th, 2008). On this day, two periods of condensational growth can be identified in Fig. 7, one during night time (from 0:00 to 7:00) and another during day time (from 12:00 to 18:00). [We would like to emphasize that we consider this particular day for the purpose of illustration how the coupled model works in the conditions close to ideal. Therefore, we deliberately choose the day when the growth due to condensation happens continuously without interruptions. The measurements were conducted under the clear sky and within the same air mass.](#)

The only free parameter in the system is the initial growth rate, taken to be 2.6 nm/h for the night, and 12 nm/h for the daytime. The time step was $\delta = 6$ min and the initial parameters for the aerosol modes were taken from the approximation of the experimental data with a lognormal distribution (night time: $N_{01} = 3130 \text{ cm}^{-3}$, $d_{p01} = 57 \text{ nm}$, $\sigma_{01} = 1.32$, $N_{02} = 165 \text{ cm}^{-3}$, $d_{p02} = 154 \text{ nm}$, $\sigma_{02} = 1.24$; daytime: $N_{01} = 3365 \text{ cm}^{-3}$, $d_{p01} = 25 \text{ nm}$, $\sigma_{01} = 1.54$, $N_{02} = 520 \text{ cm}^{-3}$, $d_{p02} = 93 \text{ nm}$, $\sigma_{02} = 1.31$).

Even such a simple model gives quite reasonable predictions of the time evolution of CS [for the time characterized by continuous growth of aerosol due to condensation](#). At night time the predicted values of the CS are higher, but from Fig. 7 it follows that the concentration of the particles decreases, which is something that we do not capture with the model in its present form. However, the model performs well for the characteristic diameter of the growing mode with the larger number concentration. During the daytime both the evolution of CS and diameter of the growing mode are predicted well assuming the constant value of $Q(t)$, but this assumption can not capture the abrupt stop of the growth of the CS in the evening.

Next, we account for the decrease in the particle number concentration during the night time in the simplest way, assuming $n_d \sim \exp(-t/\tau_{\text{loss}})$. This decay can be associated with an additional term on the r.h.s. of equation (18):

$$\frac{\partial n_d}{\partial t} + \frac{\partial(I_d n_d)}{\partial d_p} = -\frac{n_d}{\tau_{\text{loss}}}, \quad (21)$$

and the solutions (9) and (10) have the same form except for they are multiplied by the factor $\exp(-t/\tau_{\text{loss}})$. It follows then from formula (12) that the CSs are again given by the same formulas (13) and (14) simply multiplied by this factor. The results of the model calculations assuming that the number concentration of particles in the mode with a larger number concentration but smaller characteristic diameter (mode 1) is reduced by 35% and $\tau_{\text{loss}} = 7$ h (estimates obtained from experimental data by best fitting the number concentration with an exponentially decaying function) are displayed in Fig. 9 by dotted curves. The particle diameter evolution is affected very little while the CS grows significantly slower and shows now a better agreement with the measurements.

The previous example illustrates that the growth due to condensation in the atmosphere can be captured by the model. Generally, aerosol modes do not exhibit this well-pronounced continuous dynamics, with the growth process being rather often interrupted either due to the changing air mass, precipitation or some other reason, which must be well understood and parameterized before they can be incorporated into the model. However, within the time slots when the meteorological conditions are more or less stable and low-volatile organic vapours are supplied, aerosol grows due to condensation and our model can be applied.

Finally, we comment on the choice of the initial diameter of the mode 20 nm (daytime). For small particle diameters the equation for the number particle distribution should most likely include a diffusion term to account for the widening of the distribution. Starting from the smallest diameters we would end up with a non-physical narrow distribution as a result of time evolution (Lehtinen and Kulmala, 2003). Note that in the present calculations the distribution is narrowing (Fig.2) though in nature it is quite opposite. However, the precise form of a distribution seems not to be important for the CS evolution.

5 Conclusions

We have obtained a solution for the condensation equation in the range of intermediate Knudsen numbers (for particles with diameters up to ~ 500 nm). The solution is based on taking two terms of the expansion for the Fuchs-Sutugin coefficient in terms of $(1/Kn)$ at large Kn and is valid both for the constant vapour pressure and (with small modifications) for the vapour pressure changing in time.

Based on this solution, we have obtained the algebraic formulas describing the dynamics of the condensation sink (CS) in time, assuming an initial lognormal particle number-size distribution. We have tested the formulas against atmospheric observations for quasi-stationary conditions. For the typical parameters of aerosol modes in Hyytiälä (Finland), the correction results in 5.5% overestimation of CS compared with the calculations using the full Fuchs-Sutugin formula. There is also an overall error due to the approximation of the data with a lognormal distribution, which varies and can be up to 50% when the tail of the distribution corresponding to larger particles is not captured well. This error, however, does not exceed 15% when two aerosol modes are considered.

We confirm the previous results by Lehtinen et al. (2003) that CS is defined mostly by Aitken and accumulation modes with characteristic diameters ≥ 50 nm and show a diagram allowing to estimate the contribution of different modes to the CS, depending on the characteristic diameter of the mode and particle number concentration. We conclude that for typical atmospheric conditions the cluster mode with the characteristic diameter of about 2 – 3 nm and large number concentration of about 10000 cm^{-3} does not contribute significantly to CS until its geometric mean diameter grows to ≥ 20 nm.

Note that the difference between CS in the kinetic regime and CS in the intermediate regime can be estimated from Fig. 3. CS in the kinetic regime is proportional to the total surface of aerosol per unit volume. This same quantity appears in the extinction coefficient quantifying aerosol optical depth (Sundström et al., 2015). Thus, one can deduce for what parameters CS is suitable to represent aerosol impact on solar irradiance. For a typical lognormal distributions with $\sigma_0 = 1.5$, CS can be used as a proxy for extinction coefficient if an aerosol modes has a geometric mean diameter less than ~ 120 nm ($CS_{\text{cor},0}/CS_{\text{kin},0} \geq 0.8$).

Note that these parameters are typical for Hyytiälä where a strong correlation between CS and the extinction coefficient at 550 nm has been demonstrated (Virkkula et al., 2011). This is important to have in mind when choosing parameters for the quantification of biosphere-atmosphere feedback loops.

5 The differential equation for the vapour concentration was coupled with the equation for the evolution of the particle number distribution to obtain a simple self-consistent model of CS dynamics in the atmosphere. For typical atmospheric values of CS, one can use a quasi-steady state solution for the equation for the vapour concentration, as well as the analytical formula for CS can be used. This model gives reasonable results for the dynamics of CS during the periods of pronounced aerosol growth by condensation for the characteristic diameters of the mode ≥ 20 nm.

10 Note that in the framework of this model the characteristic diameter of each mode is permanently growing. In nature, in the case we considered, the growth of diameter is likely to be interrupted by the processes related to meteorology, e.g. morning and evening transitions in the boundary layer.

15 The model can be extended to investigate dynamics of a cluster/nucleation mode with a characteristic diameter of a few nm in the presence of a base mode and a time-dependent vapour concentration. As we showed, these modes will have only a negligibly small effect on the coupled dynamics of the base mode and condensing vapours while the base mode will define the condensation sink for the smallest particles. The simplest way to include the cluster/nucleation mode is to add into the system considered here a general dynamic equation with a nucleation term and a diffusion term (Seinfeld and Pandis, 2016). For larger particles, the time-dependent vapour production rate and a particle phase chemistry effect can be relevant for future investigations.

20 *Data availability.* Data measured at the SMEAR II station are available on the following website: <http://avaa.tdata.fi/web/smart/>. The data are licensed under a Creative Commons 4.0 Attribution (CC BY) license.

Competing interests. The authors declare that they have no conflict of interest.

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Parameter	Hyytiälä, 27.03.2014	Hyytiälä, 01.06.2008	Hyytiälä, 24.07.2008, 1st mode	Hyytiälä, 24.07.2008, 2nd mode
d_{p0} , nm	48 ± 7	106 ± 40	52 ± 19	132 ± 40
N_0 , cm^{-3}	2700 ± 990	2480 ± 1600	2970 ± 1240	380 ± 250
σ_0	1.62 ± 0.09	1.42 ± 0.12	1.35 ± 0.10 m/s	1.31 ± 0.11

Table 1. Parameters of the lognormal distribution for different days.

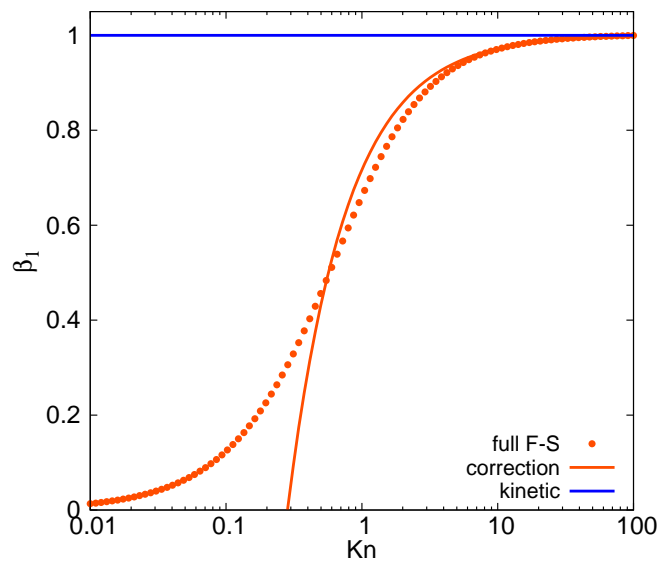


Figure 1. Fuchs-Sutugin coefficient as compared to its one term (kinetic) and two terms (correction) expansions at small $1/Kn$.

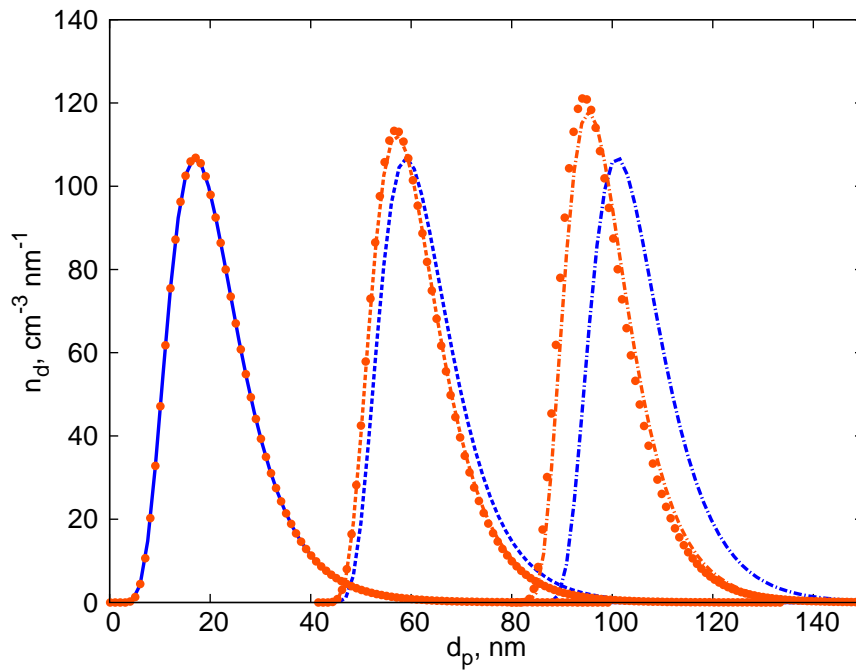


Figure 2. Time evolution of the lognormal aerosol distribution: blue - solution (9), the kinetic regime; orange - solution (10), the solution for intermediate Kn . Solid curves - initial distribution, dashed curves - after 6 hours, dash-dotted curves - after 12 hours. Orange symbols display the numerical solution of the condensation equation employing the full FS coefficient.

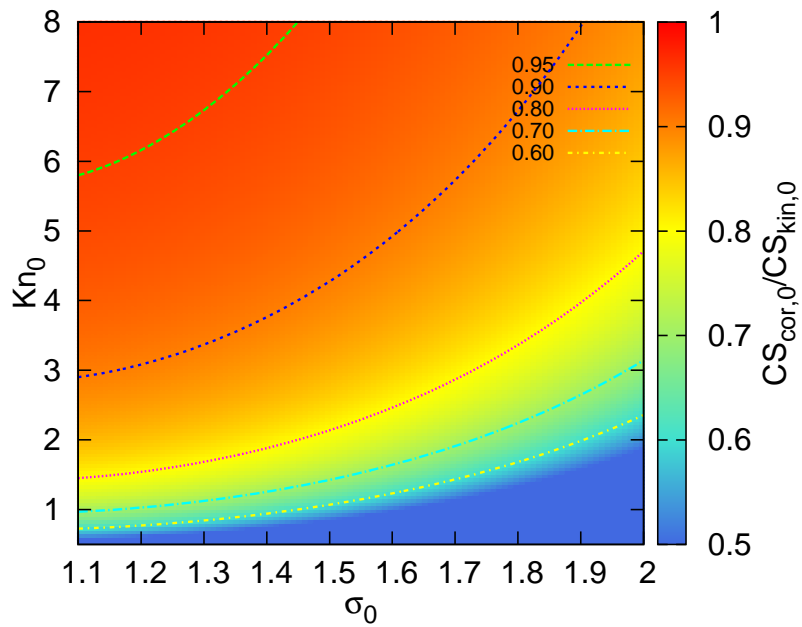


Figure 3. The ratio $CS_{cor,0}/CS_{kin,0}$ as a function of σ_0 (characterizing the width of the particle number distribution) and the Knudsen number Kn_0 (corresponding to the geometric mean diameter of the mode).

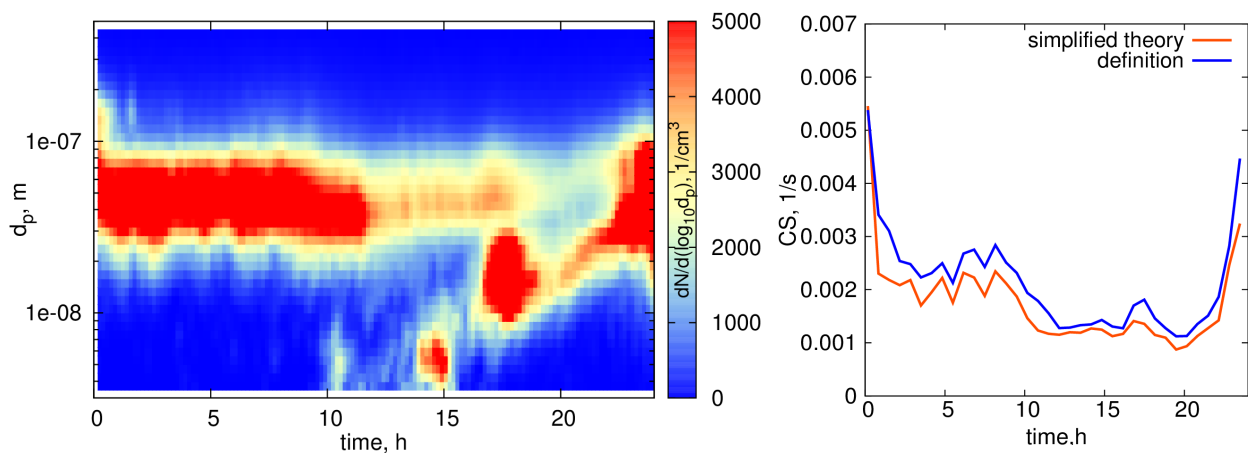


Figure 4. Left: Particle number distribution on March 27th, 2014, Hyytiälä. Right: Time evolution of the condensation sink below 500 nm. Blue curve: calculated from the definition, orange: calculated using formula (16).

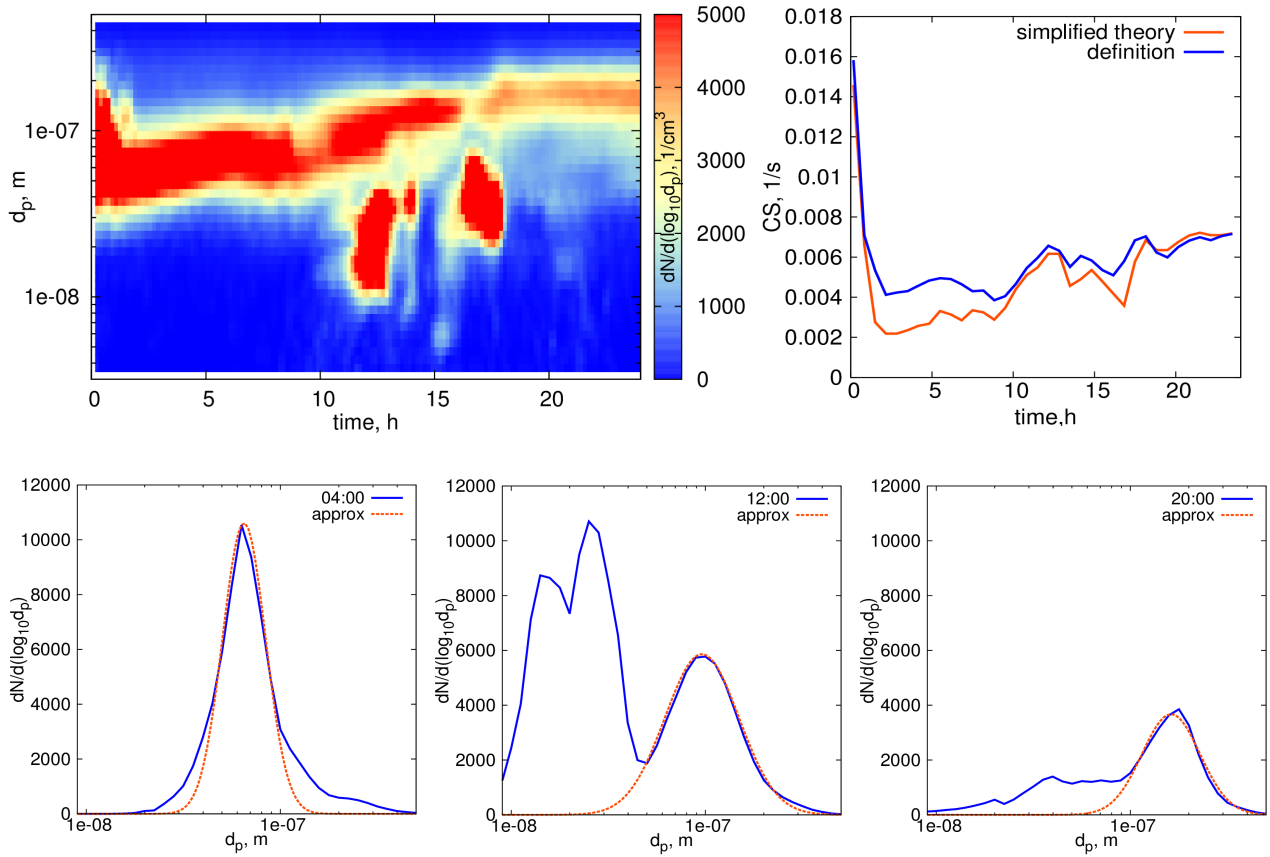


Figure 5. Upper panel: (Left) particle number distribution on June 1st, 2008, Hyttiälä. (Right) condensation sink below 500 nm, blue: calculated from the definition, orange: calculated using formula (16). Lower panel: examples of one-mode approximation of the particle number distribution.

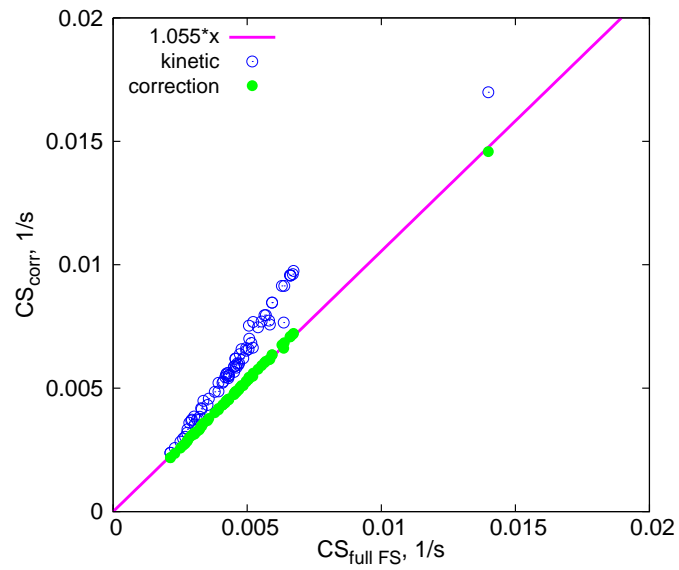


Figure 6. CSs calculated using two approximations vs CS calculated using the full FS formula. The violet line corresponds to $CS_{corr} = CS_{fullFS} * 1.055$.

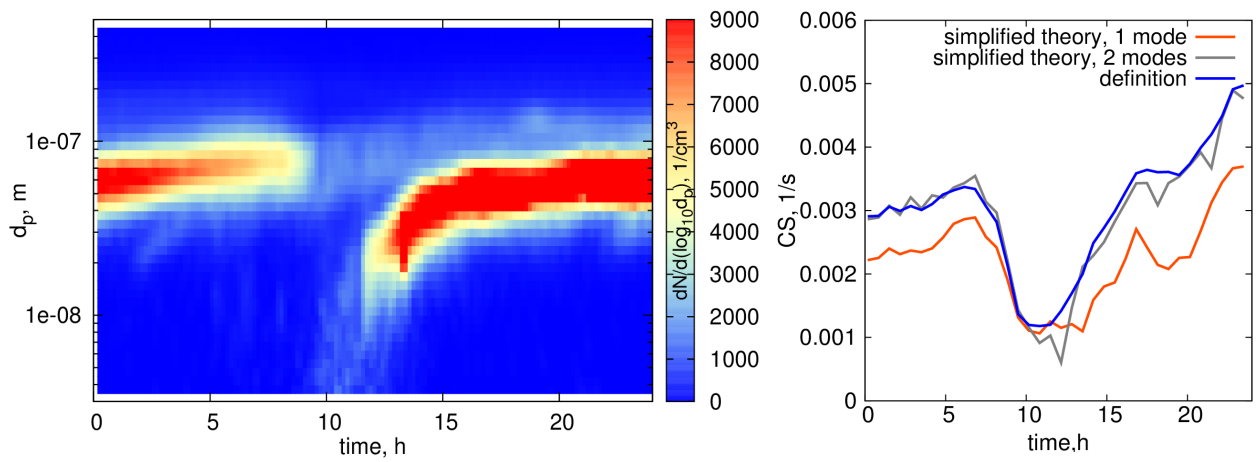


Figure 7. Left: Particle number distribution on 24th July 2008. Right: Time evolution of the condensation sink below 500 nm. Blue curve: calculated from the definition, orange curve: calculated using formula (16) for one mode, gray curve: calculated using formula (16) for two modes.

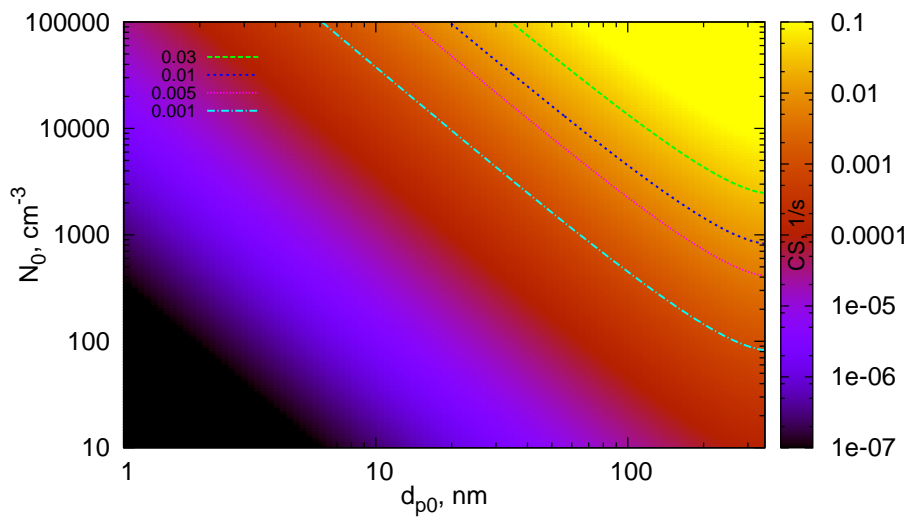


Figure 8. A diagram showing the condensation sink, CS, as a function of geometric mean diameter of the aerosol mode, d_{p0} and the particle number concentration, N_0 , for $\sigma_0 = 1.5$.

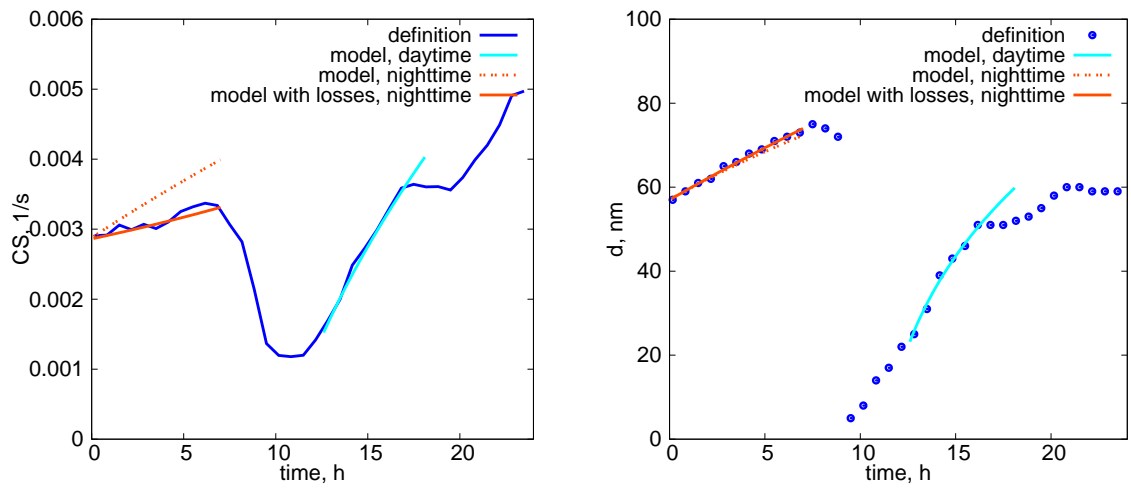


Figure 9. Left: Time evolution of the condensation sink. Blue curve: calculated from the definition, orange dashed curve: modelled with the constant particle number concentration, orange solid curve: modelled with the decreasing particle number concentration, cyan curve: modelled with the constant particle number concentration, daytime. Right: Characteristic diameter of the aerosol mode with a larger number concentration as a function of time.