Response to Referee #1

We have previously published a response to the comments of referee #1. Below we summarise how we have revised the MS in response to the refereeôs comments.

General comments:
The authors model the atmospheric boundary layer and aerosol dynamics above a pine forest in Finland, in order to examine the effect on vertical transport of aerosols, and how this impacts on the interpretation of flux measurements. They find that the vertical transport of particles within and above a forest canopy can be strongly dependent on the aerosol dynamics and measured fluxes deviate from dry deposition. This is an important result and deserves publication in ACP.

We thank the referee for encouraging comment.

I would, however, recommend proof reading the manuscript, as I found the language difficult to understand at times, meaning that the points the authors were trying to make were not always clear.
I would also like to see a description of the measurements that were used in the model.

The study relies on the model simulations and the measurements were used either to initialise the model or for evaluation of model outputs in terms of predicted particle size distributions (Fig. 1) and meteorological variables such as surface heat fluxes (Fig. 2). All other results were based on the model runs. We emphasized this more clearly in the revised MS (Introduction, last paragraph).

Description of the station and the measurements presented in the MS are described in Appendix A of the revised MS. For the input data used in the model we refer to Zhou et al. (2014) and Appendix B in the revised MS.

Minor comments:

Page 19376, line 10: ñHowever, the fact that the model is not able to reproduce the fine details of the particle formation events does not affect the generality of our results.ò Could the authors qualify this? If the modelled nucleation modes were not ñas clearò, and the particle growth overestimated, I would have thought this could have an impact on the aerosol dynamical term. Are the authors able to quantify this impact, and thus assure the reader that the same conclusions apply?

The aerosol dynamical term is dependent on the amount of condensing vapours available and the quantitative results would differ if the model was better tuned to reproduce the observed particle growth patterns. This can be seen also from our presented results where the aerosol dynamical terms differ between different days. We presented respective sensitivity analysis in the revised MS (the Supplement).

However, we meant that our qualitative conclusions were not affected by the quantitative differences between our simulations and measurements and we believe the conclusions hold.

Page 19376, line 23: ña single mode with the maximum particle amount at around 200nmò, remove ñwith the maximum particle amountò.

Removed.

Page 19377, line 13: ñThe aerosol concentration inside and above forest was homogeneous at noon and small vertical concentration gradients could not be observed from color presentation 15 in Fig. 4a.ò. This is presumably from the model results. Are there any measurements to validate this (even from published results)?

Yes, this is from model results. We addressed the comment in our earlier response. No amendments were done to the revised MS regarding this question.

Page 19377, line 18: Why ñnot shownò?

We think the respective figure does not add new information and therefore it was excluded from the MS. The figure was presented in our earlier response, which was not included in the revised MS.

Page 19378, line 3: ñnormalized to local concentrationò ï are these modelled concentrations?

Correct, clarification was included in the revised MS.

Page 19378, line 22: ñNote however that the concentration of small particles was very low in the eveningò ï will this affect the accuracy of the results?

We believe not. However, to observe/verify such a behaviour with experiments would be extremely difficult (or virtually impossible) because of large uncertainties involved in observations of particle concentrations and especially fluxes in case of small particle counts. Therefore the note was made to draw the attention to low concentration. No modification of the revised MS was done regarding this item.

Page 19381, line 11: ñexceeding deposition more than ten timesò ï do you mean exceeded by a factor of more than ten? Use this term instead, as ñmore than ten timesò could be interpreted to mean more than ten occasions.

The revised MS was corrected as suggested.

Table 1: The median values are a lot closer to unity than the averages, suggesting the latter are affected by outliers. Could the authors comment on whether this affects their conclusions?

It is true that the medians are closer to unity than the averages. This means that the averages are affected by the values deviating much from the average values. Those values are not however outliers (in terms of likely being erroneous values) but within the model context the ñtrue valuesò that correspond to certain conditions occurring in the ABL. Therefore we believe our conclusions hold. A comment was added into revised MS regarding the average and median statistics.

Page 19384, line 6: ñThe dominant condensing compounds, OH oxidation products of monoterpenes, resemble a similar profile as monoterpenes and model simulates strongest growth of nucleation mode particles at the same height.ò ï Iôm afraid I didnôt quite understand this sentence; could the authors make it a bit clearer. Also is there concentrations and/or profile data for monoterpenes? If so, please show it, or at least reference it.

The vertical profile of monoterpenes was presented in our earlier response. We have revised wording in the revised MS to be more clear.

Page 19384, line 17: ñThe concentration time change, when summed up from the surface up to the measurement level, is called the storage term and commonly accounted for in estimation of the net ecosystem exchange of carbon dioxide from the EC flux measurements.ò ï A reference would be useful here. Also correct ñin estimationò.

A reference was added Foken et al. (2012).

Figure 2: A label for the colour scale is needed for (a). Please clarify: SMEAR is the measurements?

A label to colour scale was inserted. SMEAR means Station for Measuring Ecosystem-Atmosphere Relations, a station located in Hyytiälä, Southern Finland. Explanation was added to figure caption.

Figure 3: Correct bottom axis label of (a) ñDiameterò. Where is nucleation on (b)?

Axis label corrected. The nucleation at 2 nm particle size is out of scale as explained in the figure caption. This is to improve presentation for other sizes.

Figure 6: Positive velocity means downward? Please clarify.

The positive values of the exchange velocities mean downward transport, see eq. (8). Clarification was added to figure caption.

Figure 7: There is a lot of variability with diameter in aerosol dynamics and transport timescales. Is there any measure of uncertainty?

We have answered to this comment in our earlier response. We included a new figure (Fig. 10 in the revised MS) showing the average as well as variation range of the storage change, aerosol dynamical and exchange velocities relative to deposition velocity. The figure illustrates the variability over different time moments and conditions.

Sincerely,
Üllar Rannik

Response to Referee #2

We would like to thank the referee for the valuable and encouraging comments. We agree with the referee about the over-statement of the results. However, we were excited to see the occasional strong influence of ABL and aerosol dynamics on the vertical fluxes within and above canopy. This was contrary to our expectation based on the knowledge that turbulent transfer occurs much faster than aerosol dynamical processes, enabling efficient mixing of aerosols within and above the canopy. This was an interesting observation and deserves in our opinion attention of the community working with particle flux measurements and interpretation of results. Whereas our study was based on the numerical model and a limited period of time with highly dynamical conditions, we also agree that further analysis is needed in terms of widening the parameter space as suggested by the referee. This was however not included in the scope of the current study.

Detailed response to all comments and questions follows below.

Sincerely,
Üllar Rannik

Major points:

1. Over-statement of the results: The statements about aerosol dynamics impact on particle concentrations relative to deposition (e.g. in the abstract ócan frequently exceedô) need to be clarified to note if this is FOR A given size, for NUMBER concentrations and to note in these 10 simulation days most exhibited nucleation and FOR THIS SITE . . . all these facts are rather important to the global importance attached to this statement. Making these statement more specific and tempered will not devalue the manuscript but will avoid unwarranted claims. Equally this work does not prove that óeddy covariance techniques do not generally represent dry depositionô I think it is hard to make that statement based on simulations of 10 days at a forest site in Finland. These statements in the abstract and elsewhere should be corrected. Equally the first sentence in the conclusion ï the authors have not observed ï simulations conducted using their model (which is based on some assumptions) that indicate. ...

We accept the criticism regarding the generality of results. We have performed numerical study over a pine forest in Southern Finland, with the model adjusted to respective conditions. In addition, the study period covered 10 days period when intensive aerosol formation occurred. Therefore the conclusions were revised to be applicable for the dynamical conditions presented in the study. The respective statements were revised in the MS.

2. Insufficient testing of parameter space: Given this is a numerical experiment I am unclear why the authors do not explore more parameter space. Its useful to present a brief evaluation using óreal dataô but why only do 10 days of simulations** ï the time is not really relevant surely the point is to explore the realm of plausible situations. More exploration of the parameter space should be undertaken to examine generalizability.

A period of ten days with new particle formation was chosen as the study period to analyse if and how much intensive aerosol and ABL dynamics can affect fluxes within and above canopy. For this purpose we believe 10 days is enough. We have additionally included sensitivity analysis on the saturation concentration of condensing vapors and this extends the study in terms of very important parameter driving the aerosol dynamics (presented in the Supplement). We suggest that future studies would serve useful to address wide range of conditions along with the study of the impact of canopy structure. As our study is limited to certain conditions we have revised our statements on the generality of the results (see point 1).

The following aspects of the initialization are unclear: - I understand the only data the team have access to are the 2m PSD but this is below the canopy and under weak turbulence it could be quite decoupled from the actual level where it is óappliedô i.e. at the upper boundary. This point is actually quite unclear in the manuscript but I believe a uniform PSD is applied throughout the vertical domain? (there is some (unclear) discussion on 19375. ï

Yes, uniform PSD was applied during initialisation at mid nights. The respective paragraph was amended to make it clearer.

The manuscript states the model is initialized with óvertical profiles describing the atmospheric stateô but no details are provided about what parameters are set based on which measurements. This should be clarified. ï The initialization of the gas phase chemistry is not described. Neither is the mechanism. The authors should document this fully (perhaps in Supplemental Materials)

More details of initialisation of the model were provided in Appendix B of the revised MS.

3. The authors do not provide any quantitative evaluation of the model. While this evaluation should naturally focus on the PSD, it could/should include other aspects ï e.g. physical parameters that show the turbulence conditions are being represented (the plot of TKE extends to 1500m but surely most of the órelevanceô is for much lower heights? They plot a time series of <I GUESS> surface (or maybe top of the canopy) Latent heat and sensible heat flux but these plots (Fig 2) only really serve to emphasize the diurnal cycle (which is trivial to reproduce). A scatter plot would be more meaningful as would application of statistical model skill metrics (to all measured parameters ï e.g. TKE must surely be quantified from sonics deployed at the site).

We have included in the Supplement (Fig. S1) comparison of measured and simulated TKE, H, and LE at the EC measurement level above the canopy. Whereas the heat fluxes compare with simulation results well, the ability of the model to predict TKE is fairly weak. However, we note that the present model is not able to reproduce the 3D flow at the site and from the point of view of the study it is more important to reproduce diurnal variation and dynamics of the ABL, which is mainly driven by surface heating.

4. The authors claim their results are ógeneralityô (generalizable) ï but I think they are also very specific to the canopy structure. Clear documentation should be provided of that (e.g profile of LAD), and an analysis conducted to examine the sensitivity to it.

We present information on the site in Appendix A of the revised MS. The canopy at the given site is open and we believe the aerosol dynamical effects would be more pronounced in case of dense canopies. However, more detailed study of the impact of the canopy structure remains out of the scope of this study.

5. The model is based on a number of assumption many justifiable (but maybe not full justified) assumptions. BUT it should quantify the uncertainties on their estimates of the importance of the terms they include and those they neglect.

Each modelling approach involves number of assumptions. We have utilised the model SOSAA, which is well documented in earlier publications. The model was set up for specific site and period of time involving particle formation events. Therefore the results are assumed to be applicable to similar conditions and we have acknowledged this in the revised MS. Here we point two main assumptions of the model (i) 1D model, (ii) dependence of the condensational growth of aerosol particles on the saturation vapour concentration for the lumped organic vapours. We do not have 3D model for this purpose and therefore we present the results for ideal horizontally homogeneous conditions. To test the assumption on the amount of condensing vapours, we present sensitivity simulation results in the Supplement, see Figs. S2 ï S5. Based on the sensitivity study we find that the main conclusions of the study hold.

Minor points:

1. Itôs strange to have a definitional statement (aerosol dynamics) in the abstract and even stranger when it is not linked to the previous and subsequent sentences ï I suggest rewriting the abstract to improve it.

This sentence was added during the publication phase in ACPD when clarification was asked for definition of aerosol dynamics. However, we moved the sentence from the abstract into introduction in the revised MS. The abstract was partly rewritten.

2. P19371 óleads us to the assumptions thatô I think this is really the PREMISE. I think its correct but a little bit imprecisely formulated. I would say that the vertical transport by turbulent eddies is rapid compared to other aerosol dynamics processes that impact the number particle size distribution, but as the authors suggest because other resistances to actual surface removal are slower the composite timescale for surface removal by dry deposition may under some circumstances be comparable to the time scales on which or process that act to modify the particle size distribution act. . . (i.e. we are not disagreeing but I think the formulation of the postulate could be tighter).

We modified the sentence in the revised MS.

3. The standard of English grammar is not very high ï this leads to some imprecise statements e.g.; ó The time scale of turbulent transfer is the estimate of the transfer time within turbulent air layer.ô I think perhaps a careful reading and correction would benefit the manuscript.

We have modified wording of the particular sentence in the revised MS. The MS was read and corrected, we hope it has improved.

4. P19377: The authors state óthe correspondence was not exactô (i.e. flux defined at canopy top is not = deposition) the authors say it indicates a complex relationship. . . ï This is a óthrow awayô statement ï and it is associated with a reference to Figure5a but looking at that figure I think it is still unclear how important the discrepancy is (or average and in the worst cases ï both of which should be presented) or what the cause is.

Figure 5 illustrates that the particle vertical transport at the canopy top does not exactly correspond to deposition within canopy. It can be seen that the aerosol dynamics is responsible for additional sink inside canopy for sizes up to 10 nm, creating concentration decrease as well as downward particle transport to compensate for the loss. Explanation was added into revised MS.

5. A lot of the figures are time series or plots for individual time periods ï I realize they are easy to plot but they are poor from a diagnostic perspective ï The author should generate better, more synthetic diagrams.

We have replaced Fig. 9 with a new figure quantifying the ratio of the average exchange velocity to the average deposition velocity as the time of day, as well as range of variation (as characterised by std). In addition, we have added a new figure (Figure 10 in the revised MS) presenting the same ratio as the function of particle size on three subplots for the morning, afternoon and night hours. We believe these figures help to present the large variability in the exchange velocity (relative to deposition velocity) as well as systematic deviations from one-to-one lines.

6. Figure 3 the terms (e.g. óAer. Dyn.ô) should be defined in the caption.

Definitions were added to the caption.

7. Figure 7 why is the transport óclippedô the authors should note values not shown. This figures is quite surprising on many levels ï one would not expect such large variations from Dp to Dp. . ..What does it look like when an average and sd are plotted for the entire simulation period?

The out of scale values for the transport term were included in the figure caption. Large variation with the particle size is natural for the aerosol dynamical term, which depends on the size spectrum and availability of condensing vapors. Consequently also the transport term exhibits large variation. Since the plot presents very wide range of variation (over the logarithmic scale) which evolves in time, we believe it is not reasonable to average the time scales over the entire simulation period. Alternatively, we have included in the revised MS the variation of the exchange velocity (relative to deposition velocity) with particle size, see Fig. 10 in the revised MS.

8. Figure 8 and 9 why is there such a large discrepancy in the PSD in frame (a)? ï I guess this is the model being reinitialized but it does appear to indicate the model is drifting a long way from órealityôï doesnôt that give pause to some of the more sweeping generalization statements they make in the manuscript?

The used model is a 1D model being not able to account for horizontal advection effects. This is one of the reasons (in addition to possible model limitations to reproduce the real atmospheric chemistry and aerosol physics) why the modelled PSD-s deviate with time from the measurements. The influence of horizontal advection on ABL properties, in particular to PSD, has been frequently observed at the site (see also the references in the MS and respective discussion). This is also the reason why the PSD-s were re-initialised at mid-nights. However, the re-initialisation was done by taking uniform PSD up to the previous day ABL height (the assumed residual layer) and due to initial uniform distribution the particle vertical transport during the BL growth is expected to be lower than what could occur in the actual ABL. Nevertheless, we recognise the limitation of our result in particular to dynamical conditions with new particle formation events and therefore have respectively modified our conclusions. Figure 9 was replaced with a new figure, see comment 5.

Aerosol dynamics within and above forest in relation to turbulent transport and dry deposition

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Abstract

One dimensional atmospheric boundary layer (ABL) model coupled with detailed atmospheric chemistry and aerosol dynamical model, the model SOSAA, was used to predict the ABL and detailed aerosol population (characterized by the number size distribution) time evolution. The model was applied over a period of ten days in May 2013 to~~for~~ a pine forest site in southern Finland. The period was characterized by frequent new particle formation events and simultaneous intensive aerosol transformation. ~~Throughout this study we refer to nucleation, condensational growth and coagulation as aerosol dynamical processes, i.e. the processes that govern the particle size distribution evolution.~~ The aim of the study was to analyze and quantify the role of aerosol and ABL dynamics in the vertical transport of aerosols. It was of particular interest to what extent the fluxes above canopy deviate from the particle dry deposition on the canopy foliage due to the above mentioned processes ~~from the particle dry deposition on the canopy foliage~~. The model simulations revealed that the particle concentration change due to aerosol dynamics ~~can~~ frequently exceeded the effect of particle deposition even an order of magnitude or more. The impact ~~is~~ was however strongly dependent on particle size and time. In spite of the fact that the time scale of turbulent transfer inside the canopy is much smaller than the time scales of aerosol dynamics and dry deposition~~,~~ letting to assume well mixed properties of air, the fluxes at the canopy top frequently deviated from deposition inside forest. This ~~is~~ was due to transformation of aerosol concentration throughout the ABL and resulting complicated pattern of vertical transport. Therefore we argue that the comparison of time scales of aerosol dynamics and deposition defined for the processes below the flux measurement level do not unambiguously describe the importance of aerosol dynamics for vertical transport ~~within~~ above the canopy. We

conclude that under dynamical conditions reported in the current study the micrometeorological particle flux measurements ~~such as performed by the eddy covariance technique do not generally represent~~can significantly deviate from the dry deposition into the canopy. The deviation can be systematic for certain size ranges so that the ~~conclusion applies also to~~ time averaged particle fluxes can be also biased with respect to deposition sink.

Keywords: Aerosol size distribution, aerosol and atmospheric boundary layer dynamics, turbulent transport, time scales.

1 Introduction

Turbulent fluxes of scalars are commonly measured by the eddy covariance (EC) technique above forests. From flux measurements the exchange of scalars between the ecosystem and the atmosphere is inferred by making simplifying assumptions, mainly horizontally homogeneous and stationary conditions, considering usually transport of passive scalars. From aerosol particle flux measurements deposition to ecosystem is inferred by neglecting all additional terms including the storage term. However, there are several mechanisms affecting the particle concentration, namely new particle formation, coagulation and source or sink term for a particular size resulting from condensational growth. These processes govern the particle size distribution evolution and which we refer to as the aerosol dynamical processes throughout this study. The significance of aerosol dynamical terms in comparison to dry deposition has been evaluated by comparing the respective time scales. The time scale for dry deposition for measurement level z has been estimated according to $\tau_{dep}(z) = \frac{z}{V_d}$, where $V_d = -\frac{F(z)}{C(z)}$ denotes the bulk deposition velocity defined as the ratio of the total flux divided by the concentration at the same level (Pryor and Binkowski, 2004; Pryor et al., 2013). Such a definition of the time scale of dry deposition implies that frequently the aerosol dynamical terms have similar time scales to dry deposition and therefore affect the conservation of aerosol particles concentration during the transport pathway between the EC measurement level and the collecting surfaces. Depending on the prevailing conditions i.e. the nucleation rate, the availability of condensing vapors determining the condensational growth, and the shape of the particle size spectrum, the aerosol dynamical terms can vary significantly depending on the particle size. The time scale of aerosol dynamical processes varies typically between 10^3 to 10^5 seconds (Pryor and Binkowski, 2004; Pryor et al., 2013), i.e. being on the

hourly time scale and more. This is a sufficient time to allow well-mixed conditions to establish within the unstable day-time ABL, where the mixing time scale is estimated to be around 10 minutes (e.g. Stull, 1988). Under near-neutral and stable conditions such efficient mixing throughout atmospheric column cannot be assumed. Instead the characteristic time scales of turbulent transfer within and above forests have been estimated by different approaches (e.g. Zelger et al., 1997; Rinne et al. 2000, 2012, Rannik et al., 2009b). Such time scales of turbulent transfer depend on the observation conditions but typically remain in the order from a few tens of seconds to a few hundreds of seconds. In spite of different definitions used and large variation range of the time scales characterizing the scalar transport between the observation level and the collecting surfaces within forest, the turbulent transfer can be expected to occur much faster than the aerosol dynamical processes.

The aerosol particle dry deposition is strongly size-dependent as different mechanisms operate at different particle sizes. Respectively, the time scale of dry deposition depends on particle size and exhibits its maximum at around 100 nm. For small particles with a few nm in diameter this dry deposition time scale can be orders of magnitudes smaller due to efficient removal mechanism by Brownian diffusion. At particle sizes larger than 100 nm the particle collection is again enhanced due to interception and inertial impaction mechanisms (Petroff et al., 2008) and the respective time scale of dry deposition is smaller. In general, the dry deposition time scale has been frequently estimated to be in the same order of magnitude as the time scale for aerosol dynamics, leading to a conclusion that flux divergence may occur during transport due to aerosol dynamics (Pryor and Binkowski, 2004; Pryor et al., 2013).

The time scales of turbulent transfer and the time scale of dry deposition embed essentially different definitions and can lead also to different conclusions about the significance of aerosol dynamical terms during the transport between the underlying surfaces and the measurement level. The time scale of turbulent transfer is the ~~estimate~~ characteristic time of the transfer ~~time~~ within turbulent air layer. Dry deposition includes in addition the transport pathway within the laminar air layer surrounding the collecting surfaces. In the resistances framework (e.g. Monteith and Unsworth, 1990), the dry deposition includes the aerodynamic (corresponding to turbulent transport) as well as the leaf laminar sublayer resistances and under most conditions the dry deposition is limited by the laminar boundary layer transfer (e.g. Petroff and Zhang, 2010). Therefore comparison of the time scales of turbulent transport and dry deposition with that of aerosol dynamics leads us to the assumptions that (i) turbulent

transport within and above forest is relatively fast and no significant transformation of aerosol population occurs within the respective time scale, and (ii) depending on particle size the removal of aerosols via dry deposition occurs at the comparable time scale with aerosol dynamics and therefore the aerosol population ~~is~~ can be modified during the removal process. Such modification occurs on hourly time scale and therefore is expected to occur throughout the ABL, where aerosol dynamical processes can depend strongly on height within the ABL via vertical profiles of condensing vapors.

The purpose of this study is to analyze the magnitude of different terms in the particle number conservation equation and to evaluate the time scales of particle turbulent transfer, aerosol dynamical processes and dry deposition over wide range of particle sizes. Further, we evaluate the effect of these terms on inferring particle deposition velocities from flux measurements by micrometeorological techniques, in particular the influence on estimation of functional dependencies as well as systematic biasing effects. The study relies on the simulations by the model SOSAA and the measurements were used only to initialise the model (see Section 2.2 and Appendix B) or for evaluation of model outputs in terms of predicted particle size distributions and meteorological variables such as heat fluxes above canopy (see Section 3 below). Non-stationary conditions will be considered by simulating detailed ABL and aerosol dynamics inside and above the forest canopy during a period of 10 days, which includes highly dynamical conditions with new particle formation.

2 Materials and methods

The model was set up for a pine forest site in southern Finland and initialized with available measurements performed at the SMEAR II station. For description of the site and initialisation of the model see Appendices A and B, respectively. The analysis relies on evaluation of the significance of different terms of the particle conservation equation.

2.1 Conservation equation for aerosol size distribution

In horizontally homogeneous conditions, neglecting molecular diffusivity and applying the first order closure to turbulent flux

$$\overline{w'n'} = -D_t \frac{\partial \bar{n}}{\partial z}, \qquad (1)$$

the conservation equation for time-averaged particle number density $\bar{n} = \frac{d\bar{N}}{d\log_{10} D_p}$ inside the canopy can be written as

$$\frac{\partial \bar{n}}{\partial t} + \frac{\partial}{\partial z}\left(- D_t(z)\frac{\partial \bar{n}}{\partial z} - w_s \bar{n} \right) = -a(z) v_c \bar{n} + S_{ad}, \qquad (2)$$

where $\bar{N}$ is the average particle number concentration, D_p the particle diameter, D_t the particle turbulent diffusivity, w_s the settling velocity, v_c the particle collection velocity by vegetation, and a denotes the all-sided leaf area density. The source/sink term S_{ad} incorporates all aerosol dynamical terms, consisting of nucleation S_{nucl}, condensational growth S_{cond} and coagulation S_{coag} terms. If the condensational growth rate is considered as $I_{cond}(\log_{10} D_p) = \frac{d\log_{10} D_p}{dt}$, then the respective source/sink term in Eq. (2) is expressed as $S_{cond} = \left[\frac{\partial \bar{n}}{\partial t}\right]_{cond} = -\frac{\partial\left(I_{cond}\bar{n}\right)}{\partial \log_{10} D_p}$. For particle size range up to a few micrometers D_t can be assumed to be equal to the eddy viscosity of the flow. The settling velocity w_s is given as

$$w_s = \frac{C_c g \rho_p D_p^2}{18\eta}, \qquad (3)$$

where g is the acceleration due to the gravity, η the dynamic viscosity of air, ρ_p the particle density, and C_c the Cunningham slip correction factor (e.g. Hinds, 1982).

For the comparison of the significance of different terms of the conservation equation, the Eq. (2) was re-written so that the sum of all terms equaled zero, and the transport due to settling was merged with the particle collection by vegetation as

$$\left[-\frac{\partial \bar{n}}{\partial t}\right] + \left[\frac{\partial}{\partial z}\left(D_t(z)\frac{\partial \bar{n}}{\partial z}\right)\right] + \left[-a(z)v_c\bar{n} + \frac{\partial}{\partial z}\left(w_s\bar{n}\right)\right] + \left[S_{ad}\right] = 0, \qquad (4)$$

where the terms were called consequently as the storage, the (vertical) transport, the particle deposition and the aerosol dynamical terms. Further, integration of Eq. (4) from the forest floor surface up to the canopy top h was used to define the change velocities in analogy to deposition velocity. The change velocity due to particle deposition was defined as

$$V_{dep} = \frac{1}{\bar{n}(h)}\int_0^h \left[-a(z)v_c\bar{n} + \frac{\partial}{\partial z}\left(w_s\bar{n}\right)\right]dz \qquad (5)$$

and the change velocity due to aerosol dynamics as

$$V_{ad} = \frac{1}{\bar{n}(h)} \int_0^h S_{ad}\, dz . \quad (6)$$

In particular, for the transport term the respective change velocity was defined as

$$V_{transp} = \frac{1}{\bar{n}(h)} \int_0^h \frac{\partial}{\partial z}\left(-\overline{w'n'}\right) dz = -\frac{\overline{w'n'}(h) - \overline{w'n'}(0)}{\bar{n}(h)} . \quad (7)$$

Note that in the modelling approach the vertical flux at the canopy top was obtained from the gradient diffusion approximation (1) and the flux at the surface was defined by the ground deposition parameterization, which was applied as the sink term in the lowest model layer. Therefore in our model calculations $\overline{w'n'}(0) = 0$ and the transport velocity equaled to the exchange velocity defined at the canopy top by

$$V_e = -\frac{F(h)}{\bar{n}(h)} . \quad (8)$$

The time scales of the processes affecting the particle concentration inside the canopy were defined by

$$\tau = \frac{h}{V} , \quad (9)$$

with the change velocities V_{dep}, V_{ad} and V_e defining the time scales for deposition τ_{dep}, aerosol dynamics τ_{ad} and exchange τ_e, respectively. These time scales were calculated based on the numerical modelling results by SOSAA.

2.2 Simulation of aerosol transport and dynamics by model SOSAA

The model to Simulate the concentration of Organic vapours, Sulphuric Acid and Aerosols (SOSAA) is a 1.5 order RANS (Raynolds Averaged Navier Stokes) model SCADIS (SCAlar DIStribution, 1D version, Sogachev et al., 2002, 2012) coupled with detailed biogenic emissions, chemistry and aerosol dynamics. SCADIS describes the exchange between the vegetative canopy and atmosphere by considering the vegetation as a multi-layer medium and implementing parameterizations for radiation transfer, drag forces on leaves, and stomatal conductance. The particle deposition processes in SOSAA are treated in the same manner as in the study by Lauros et al. (2011) based on the parameterization by Petroff et al. (2008). The parameterization considers Brownian diffusion and takes into account the influence of leaves on particle interception, impaction and settling. The model has been applied extensively in different forest sites for various studies concerning biogenic emissions, chemistry and aerosol formation (e.g. Kúrten et al., 2011; Boy et al., 2013; Smolander et al., 2014; Mogensen et al.,

2015; Zhou et al., 2015). Detailed model description is presented by Boy et al. (2011) and Zhou et al. (2014).

The model set-up in this study was the same as in the study by Zhou et al. (2014) except that only kinetic nucleation mechanism was employed in aerosol dynamics simulation (Weber et al., 1997; see also Sect. S2 in the Supplement). Zhou et al. (2014) presented the ability of SOSAA to reconstruct new particle formation events at Hyytiälä, which was the same site as in this study. The model was initialised with vertical profiles describing the initial atmospheric state (see Appendix B) and aerosol size spectrum observed at the surface, and run for 10 days time period similarly to Lauros et al. (2011). The aerosol size distribution was initialised each day at 0:00 LT based on the measurements at 2 m height. The first day the concentration profile was assumed constant (the same as at 2 m height) up to determined night time Stable Boundary Layer (SBL) height (320 m) and 10% of the concentration values within the SBL above this level. During the next days the concentration profile was taken constant as per measurements at 2 m level up to the maximum ABL height occurring during the previous day and 10% of the within SBL values above that level. The initialisation during the first day corresponded to the conditions of horizontal advection with very different properties of the air above the SBL, whereas during the other days the night time residual layer was assumed to retain the same properties as the SBL. The implications of these two contrasting assumptions for ABL mixing and vertical transport of aerosols will be discussed in Sect. 3.4. For meteorology simulations 10 sec time step was used along with the explicit forward in time integration method. The aerosol dynamics was simulated with 60 sec time step.

2.3 Lagrangian estimation of turbulent transfer time

The Lagrangian stochastic (LS) simulations were used to estimate the turbulent transfer time. The conventional approach of using a LS model is to release particles at the surface point source and track their trajectories towards the point of interest forward in time (e.g., Wilson and Sawford, 1996). In case of horizontally homogeneous and stationary turbulence, the mean Lagrangian turbulent transfer time at the canopy top due to a sustained source located at height z_0 (near forest floor) can be described as

$$\tau_L(z) = \frac{1}{N}\sum_{i=1}^{N}\tau_i \,, \qquad (10)$$

where τ_i denotes the travel time of trajectory *i* at the moment of intersection with observation height. For LS modelling the turbulence statistics such as the turbulent kinetic energy (TKE) and the vertical eddy diffusivity obtained from SOSAA were used to define the turbulent profiles of the dissipation rate of TKE and variances of the wind speed components.

3 Results

The selected time period consisted of 10 days in May 2013, day of year (DOY) 121 (01 May) to 130 (10 May). On several days clear particle formation patterns were observed at the smallest particle sizes around mid-day, with subsequent growth to larger particle sizes (Fig. 1). In all days significant aerosol dynamics was taking place in terms of particle growth. The model simulations reproduced the observed particle size distributions qualitatively, however being not able to reproduce the exact particle size distribution patterns. In particular, during days with new particle formation the observed nucleation modes were not as clear; also the particle growth was overestimated, which can be observed clearly during the second half of the period. With respect to condensational growth of aerosols and resulting patterns of aerosol particle distributions a sensitivity analysis was performed (Fig. S2 in the Supplement). The results of the sensitivity analysis are summarized in the end of Sect. 3. ~~However, the fact that the model is not able to reproduce the fine details of the particle formation events does not affect the generality of our results.~~

The ABL height varied between about 600 (DOY 130) and 1400 m (DOY 123) as the peak height during different days (Fig. 2a). The heat fluxes were the primary drivers of the ABL growth and buoyancy driven TKE. The simulated latent and sensible heat fluxes corresponded well to those measured at the site (Fig. 2b,c), but the simulated TKE had weak correlation with the values observed above the canopy (Fig. S1 in the Supplement). We ascribe this to the limited ability of the 1D model to reproduce the actual flow field at the site. However, for the current study it is more important to reproduce diurnal variation and dynamics of the ABL, which is mainly driven by surface heating. The selected ten days period showed significant variability in terms of aerosol and ABL dynamics and was therefore selected as the study case.

3.1 Aerosol dynamics and transport inside and above forest

The particle conservation terms were evaluated inside forest at 07 May (DOY 127), 12:00 and 21:00 LT (UTC+2 h). At noon the particle size spectrum was bi-modal, with nucleation and larger particle modes, by evening the nucleation mode had grown and almost merged into a single mode ~~with the maximum particle amount~~ at around 200 nm (Fig. 3a). The rate of change by each term (as defined by the terms in Eq. 4) showed large particle sink due to deposition, which was compensated by the transport term at noon (Fig. 3b). The aerosol dynamical term was dominated by the condensational growth term, except at sizes smaller than a few tens of nm where coagulation was also important and at smallest sizes were particles due to nucleation appeared. The aerosol dynamics reduced the particle number of small particles less than about 10 nm in diameter, adding respectively particle counts at larger sizes. The aerosol dynamical terms were reflected in relatively similar pattern in particle storage change (defined by the first term of Eq. 4). The positive value of the storage term implies decrease of particle concentration and negative increase, respectively. In the evening at 21:00 LT the change rates of small particles (less than 20 nm) were small due to low particle counts in this part of the size spectrum (Fig. 3a). The similarity (in magnitude, but opposite in sign) of aerosol deposition vs. transport and aerosol dynamical vs. storage change terms held also in the evening, letting to conclude that particle loss due to deposition was mainly compensated by vertical transport and aerosol dynamical processes modified the concentration in time.

The aerosol concentration inside and above forest was homogeneous at noon and small vertical concentration gradients could not be observed from color presentation in Fig. 4a. The deposition pattern (dependence on particle size and height) was again similar to transport patterns (Figs. 4d and c). Aerosol dynamics affected the number concentration similarly throughout the column as presented in Figs. 4e and b. The same qualitative conclusions held also for the evening time 21:00 LT (not shown).

When integrating the terms of the conservation equation (Eq. 4) from the surface up to the canopy top and normalizing with the concentration at the canopy top, one obtains change velocities as defined in Sect. 2.1. Such change velocities are comparable with the deposition velocity or the exchange velocity, which can be experimentally obtained from the flux measurements above the canopy. In terms of change velocities the deposition velocity (defined by Eq. 5) and the transport velocity (defined by Eq. 7 and being equivalent to the

exchange velocity in Eq. 8) appeared near symmetric for all particle sizes at noon (Fig. 5a). However, the correspondence was not exact, meaning that the flux defined at the canopy top did not correspond exactly to particle deposition. This was due to aerosol dynamics being responsible for additional sink inside the canopy for sizes up to 10 nm, creating concentration decrease as well as additional downward particle transport to compensate for the loss. Much larger differences in the respective patterns were observed in the evening at 21:00 LT, especially at small particle sizes (Fig. 5b). This implied a more complex relationship between particle source sink/terms (deposition and aerosol dynamics) and vertical mixing.

The vertical profiles of the aerosol dynamical term (normalized to simulated local concentrations, defining the local change rates) and the particle vertical fluxes (normalized with local concentration, defining the local exchange velocity) differed significantly for particle sizes and time of day (12:00 LT compared to 21:00 LT 07 May), Fig. 6 upper and lower panels. The respective ABL heights were approximately 710 and 510 m according to the model results. At noon the particle deposition and aerosol dynamics led to vertical particle transport that depended on particle size and height. In the lower part of the ABL the small particles (3 and 10 nm) were transported downward to compensate for deposition sink inside forest and particle loss through aerosol dynamics. The 100 nm particles were transported downward throughout the atmospheric column. For particles of 30 nm and 300 nm size it was predominantly the aerosol dynamics that drove the vertical transport, leading mostly to upward particle flux at heights above forest. The particle concentration gradients (Fig. 6a/u) were consistent with the exchange velocities. In the evening, when the vertical transport was more limited due to moderately stable conditions (the Obukhov length defined by the fluxes at the canopy top being L = +130 m), the vertical profiles showed even more complex pattern (Fig. 6 lower panels). Particles with 3 and 10 nm in diameter were transported downward up to about 50 to 100 m height (to compensate for the loss inside the canopy), whereas above these heights up to about 500 m upward flux occurred to compensate for aerosol dynamical loss in the higher part of the atmospheric column. Note however that the concentration of small particles was very low in the evening (Fig. 6a/l). The larger particle sizes (300 nm) were little affected by the aerosol dynamics in the evening and downward transport occurred (in contrast to noon). Figure 6 illustrates complex dynamics between the aerosol sources and sinks and transport in the atmospheric column, leading to aerosol dynamical term and vertical exchange that can differ in sign as a function of height for a certain particle size (for example for 10 nm particles at 12:00 and 21:00 LT).

3.2 Time scales of processes

The importance of aerosol dynamics on particle exchange measurements has been frequently assessed by comparing the time scales of aerosol dynamical and transport processes. Figure 7 presents the time scales defined in Sect. 2.1 and compares those with the Lagrangian turbulent transfer time scale determined according to Sect. 2.3. The time of turbulent transfer within forest (simulated as the time for an air parcel to travel between the surface and the forest height) was mostly much shorter than the time scales of deposition and aerosol dynamics. Only at smallest particle sizes and stable conditions the turbulent time scale became comparable to the time scales of particle deposition and aerosol dynamics (Fig. 7b). The transport time scale, defined by Eqs. (9) and (7), accounts also for the effect of sources and sinks inside the canopy and is therefore very different from the turbulent transfer time scale τ_L. The transport time scale was determined mainly by deposition and modified by the impact of aerosol dynamics, reflecting the fact that particle vertical transport is mostly controlled by the sources and sinks and being not limited by turbulent transfer speed.

The time scale of particle deposition strongly depended on particle size (resulting of respective dependence of particle collection on particle size), whereas the time scale of aerosol dynamics was occasionally shorter than the deposition time scale (even an order of magnitude, depending on particle size). Even though the turbulent transfer time scale τ_L was much shorter than the other time scales, the flux at the canopy top deviated from the deposition to vegetation elements (can be inferred from the comparison of the deposition and the transport time scales). Note that even the sign of the flux at the canopy top differed for particles of about 100 to 300 nm in diameter, see the sign of the transport time scale in Fig. 7a. Although very short turbulent transfer time would suggest fast and efficient mixing (and therefore correspondence of flux to deposition), the difference can be explained by the importance of the aerosol dynamics which affects the concentrations throughout the atmospheric column and therefore drives the vertical redistribution of particles via vertical transport.

3.3 Time evolution and statistics of particle exchange

The idea behind micrometeorological particle flux measurements is to determine the particle dry deposition fluxes or equivalently the deposition velocities. Thus it is assumed that the fluxes observed above forest represent the deposition fluxes. Figure 8 compares the change

velocities defined in 2.1 to the respective deposition change velocities during the first day of the simulations 01 May (DOY 121) and a following nucleation day 02 May 2013 (DOY 122). These two days differ in terms of initialization of vertical aerosol profiles at midnight (see Sect. 2.2). During the first day the aerosol dynamics affected little the particle concentrations inside forest, but 100 and 300 nm sizes were affected strongly by vertical transport occurring during the mixed layer (ML) growth period prior to noon. The initial concentration profile during this day corresponded to the conditions of horizontal advection. During the second day the aerosol dynamical term exceeded the deposition term several times (Fig. 8c). Respectively, the storage change varied approximately in the same limits, being opposite in phase (Fig. 8b). The variation of the exchange velocity with respect to deposition was smaller (Fig. 8d), consistently with the analysis of Fig. 3 where the vertical transport was the main mechanism compensating for aerosol loss due to deposition. Nevertheless, also the magnitude of the exchange velocity can differ several times compared to that of deposition. During the new particle formation and ABL growth period of the second day the vertical particle exchange showed downward transport of small particles (3, 10 and 30 nm) and upward transport of 100 nm particles. In particular during the first day (DOY 121), the upward particle transport was synchronous with the storage change i.e. the concentration decrease (Fig. 8b) referring to the dilution of concentration within the canopy. Downward transport of 10 nm particles during the second day in turn exceeded significantly the particle deposition. This particle size range was affected then by changing (from negative to positive) aerosol dynamical term during the morning hours due to particle growth (Fig. 8c), which was due to the fact that 10 nm size was on the lower edge of the dominant mode of the particle size spectrum (Fig. 8a). Note also that the storage change of 10 nm particles was similar to the aerosol dynamical term (opposite in sign) and not to the exchange velocity. Therefore the relatively large downward flux during the second day (DOY 122) was mainly driven by the aerosol dynamics occurring at night, whereas the growth of the ML initiated strong vertical mixing.

~~May 7th 2013 (DOY 127) was a day with different meteorological conditions compared to May 1st and 2nd (DOY 121 and 121). During the night turbulence was suppressed resulting in very low ABL during the preceding night (Fig. 2a). During this day the night-time aerosol size spectrum peaked at larger sizes (Fig. 9a) compared to May 2nd (Fig. 8a). Respectively, the 30 nm particle size was affected strongest by the aerosol dynamics (Fig. 9c) and the size experienced very large downward transport velocity (Fig. 9d), exceeding deposition more~~

~~than ten times. It is only at 9 am when the dilution of concentration was observed systematically for 300 nm sizes (Fig. 9b) presumably occurring due to ABL growth occurring later in this day, coinciding with upward particle transport (Fig. 9d). The effect was however relatively weak due to the assumption of missing horizontal advection above the SBL at night. During the following night (preceding to DOY 128) the aerosol dynamical processes were less effective, whereas the next night (preceding to DOY 129) the aerosol dynamical processes affected the aerosol population very strongly at all sizes except 3 nm. DOY 127 and 129 were the days with preceding low SBL heights and correspondingly limited vertical mixing in contrast to DOY 128, referring to the enhanced role of aerosol dynamics at stable nights.~~

In order to understand overall trends and variability in aerosol dynamics and transport, the diurnal patterns of the averages together with the range of variation were presented in Fig. 9 for three particle sizes characterizing the nucleation (10 nm), Aitken (50 nm) and accumulation (300 nm) modes. For 50 nm particles the aerosol dynamics was a sink at nights, whereas the condensational growth served as the source of 300 nm particles round the clock. The variation range of the aerosol dynamical term can be very large indicating the role of ABL development during different days. Whereas the variation range of the aerosol dynamics and storage was large generally at nights, the vertical exchange deviated from deposition mainly during the early morning SBL and further ABL growth period till noon (Fig. 9c). During this period the 50 nm particle fluxes were larger than induced by deposition, and during the ABL growth the 300 nm particle fluxes were lower than would have corresponded to deposition, on the average.

Further we looked how different particle sizes were affected during different stages of the ABL state. At night the aerosol dynamics affected wide range of particles and performed as the sink for particles less than 100 nm and source for larger particles, on the average (Fig. 10c). The aerosol dynamical sink/source led primarily to particle concentration change. During the morning hours from sunrise till noon the ABL growth induced enhanced downward transport of about 30 nm to 200 nm particles, whereas vertical downward transport of larger particles was less than deposition sink (Fig. 10a). During the afternoon all the change velocities exhibited less variation compared to morning and night hours. Consequently deposition was also the best represented by the averages fluxes at the canopy top in the afternoon, with biggest deviation coinciding with the minimum in deposition velocity at around 100 nm (Fig. 10b). Figures 8, 9 and 10 (see also Fig. S3 in the Supplement) illustrate

that both the aerosol dynamics and ABL growth can strongly affect the vertical transport of aerosols and the fluxes above the canopy can deviate significantly from the deposition occurring within the canopy.

Due to instrumental limitations or by intention (frequently to obtain statistically significant particle counts in order to reduce particle flux random errors) a certain size interval of particles is measured. Fig. ~~10~~ 11 presents the ~~simulated~~ vertical exchange ~~velocities~~ velocity size integrated values to represent the nucleation (3-30 nm), Aitken (30-100 nm) and accumulation (100 ï 1000 nm) mode particles. During the first day with assumed conditions of horizontal advection the size-integrated particle fluxes showed clear upward transport during the morning hours for 30-100 and 100-1000 nm size ranges. The same has also been observed from the measurements and interpreted as the upward transport due to ABL growth and resulting dilution of relatively particle-rich air within forest with the particle-poor air transported down from aloft (e.g. Nilsson et al., 2001). The days with very large (both positive and negative) values of the exchange velocities compared to deposition velocities corresponded to the days with preceding very low ABL heights at nights (DOY 127, 129, 130). Therefore the ABL development can be identified as one of the main reasons for the large variation in vertical transport of particles. In case of experimental flux measurements the statistical uncertainty as well as natural variation originating from spatial heterogeneity and horizontal advection can additionally contribute to the variance of the calculated fluxes, leading to flux patterns with large variation, being often difficult to interpret.

Table 1 presents the statistics of the fluxes at the canopy top (relative to deposition) for different particle sizes. Whereas for smaller particles 3-10 the time-average particle flux statistics converged to particle deposition within forest, for larger particles the fluxes (if measured by the micrometeorological technique) ~~would be~~were biased in representing the particle deposition even on the average. The largest deviations of the particle fluxes from dry deposition sink occurred during the morning period when most intensive aerosol dynamics and ABL development took place (Table 2). Consistently with Fig. 10b at that time 30-100 nm downward particle fluxes exceeded dry deposition and in the size range 100-1000 nm the downward fluxes accounted for approximately half of the deposition sink.

Finally, we performed sensitivity analysis of our simulations with respect to saturation concentration of condensing vapors, which affects the condensational growth of aerosols. Two additional cases with low saturation vapor concentration (equivalent to more

condensation) and high saturation vapor concentration (equivalent to less condensation) were tested (Sect. S2 in the Supplement). Whereas the high saturation vapor concentration case led to less apparent nucleation mode in the particle size spectrum, the low saturation vapor concentration implied more pronounced and clear particle growth patterns during the nucleation days (Fig. S2 in the Supplement). The storage change, aerosol dynamics and exchange velocities were studied for given scenarios (Figs. S3 to S5 in the Supplement). The main difference observed was that in case of high saturation vapor concentration, due to slower growth of particles, the effect of aerosol dynamics persisted longer in the morning and affected the Aitken mode particles as represented by 50 nm (Fig. S4c in the Supplement) along with similar impact on exchange velocity that overestimated dry deposition for given particle size (Fig. S5c in the Supplement). However, as revealed by the sensitivity analysis of different scenarios, the overall qualitative behavior was not significantly different.

3.4 Discussion of results

3.4.1 Aerosol dynamics and deposition

~~We have observed~~The simulations have shown that aerosol dynamics can have significant impact on aerosol population depending on particle sizes. It is mainly the condensational growth that can increase or decrease the particle numbers at certain sizes depending on the shape of the particle size spectrum. The aerosol dynamical impact on particle concentration at certain sizes can be equal to or even significantly exceed in magnitude the particle loss due to deposition within the canopy. This is in particular true for particle sizes at which deposition rate is minimal. Consistently with our result, Pryor and Binkowski (2004) and Pryor et al. (2013) have found that frequently the time scales corresponding to particle deposition and aerosol dynamical processes are in the same order of magnitude and therefore induce the concentration change with comparable magnitude. Pryor et al. (2013) evaluated these time scales to be in the order of 1 to 10 hours during the daytime in summer over a pine forest. In the current study we presented that the aerosol dynamical time scale can be from approximately half an hour to tens of hours.

The time scales of turbulent transfer and vertical transport were determined to be essentially different. The vertical transport of aerosols was limited by the deposition and aerosol dynamical processes and only at stable conditions the turbulent transfer could become limiting to vertical transport of particles. The turbulent transfer time scales estimated in the current study by using the LS trajectory simulations were in the order of minutes during the

day-time and could be up to a few tens of minutes under SBL conditions. Some other definitions of the time scales have been used in the analysis of the significance of chemical transformation of reactive scalars during transport pathway between the measurement level and sources or sinks located primarily at leaf surface. Rinne et al. (2000, 2012) used the ratio of the observation height to the friction velocity as the estimate for the mixing time scale. Zelger et al. (1997) used the definitions of Eulerian and Lagrangian turbulent time scales to characterize the turbulent transfer within and above forest. Holzinger et al. (2005) instead used the estimate of the residence time and obtained the value about 1.5 minutes for day-time conditions. The Lagrangian turbulent transfer times obtained in this study were consistent with the previous studies including the time scales obtained by the same approach by Rannik et al. (2009b).

3.4.2 Dynamics within ABL

The times scales of aerosol deposition and dynamics are much longer than the turbulent transfer times within the forest canopy. Therefore, one would expect a minor impact of aerosol dynamics on particle population during the vertical transfer within forest under most of the observation conditions and a relatively good vertical mixing of aerosols within and above forest. Nevertheless, we have seen in the current study that the vertical fluxes at the canopy top can deviate significantly from what would be expected from dry deposition only. From current model simulations we have seen that the aerosol dynamics is an important mechanism of aerosol transformation throughout the ABL, whereas the aerosol deposition occurs only inside the forest canopy. In addition, the impact of aerosol dynamics is height dependent. ~~At levels close to~~Within the canopy the emissions of the precursor gases for particle condensational growth (the volatile organic compounds) occur. The dominant condensing compounds are the~~,~~ OH oxidation products of monoterpenes, which form during the transport pathway from inside forest to higher levels in the ABL~~, resemble a similar profile as monoterpenes and model simulates strongest growth of nucleation mode particles at the same height~~. The concentrations of the condensing compounds are therefore larger within and immediately above the canopy and decrease with height. Such height dependence of the condensational growth of particles can lead to modification of concentration gradient and vertical flux profile. Even though the atmospheric mixing is fast compared to above discussed processes, we believe it is the extensive source-sink term by aerosol dynamics that operates throughout the atmospheric column (compared to the impact of deposition inside the canopy

only) and can thus create significant vertical flux divergence and even upward particle transport.

The concentration time change, when summed up from the surface up to the measurement level, is called the storage term and commonly accounted for in estimation of the net ecosystem exchange of carbon dioxide from the EC flux measurements (e.g. Foken et al., 2012). Such approach inherently assumes that the storage change results from the source/sink activity below the observation level. Rannik et al. (2009a) studied the relevance of the storage term in estimation of the dry deposition from particle flux measurements. They concluded that in case of aerosol particles the relevance of the storage term could not be established because of the different physical reasons for the concentration change during different phases of diurnal development of the ABL. This study supports the conclusion with the observation that the particle concentration change is primarily in correlation with the aerosol dynamics and the change occurs throughout the ABL. Therefore the particle storage change (which corresponds to accumulation or depletion) is not in general the sole component of the particle conservation equation that could help to improve particle deposition estimation from the flux measurements carried out above forest.

3.4.3 Upward particle fluxes

Particle fluxes determined by the micrometeorological techniques show typically large variability in magnitude as well as in sign. Occurrence of upward particle fluxes has been frequently reported in the literature (Pryor et al., 2007; Grönholm et al., 2007; Whitehead et al., 2010; Pryor et al., 2013). Even after careful classification of observations according to wind direction in order to remove the cases possibly affected by anthropogenic emissions, flux observation analyses by Pryor et al. (2008) revealed significant fraction of observations indicating emission. The upward particle flux values can be the result of large random uncertainty or caused by upward particle transport due to physical processes. Random flux errors of particle fluxes are due to stochastic nature of turbulence, instrumental noise, and (limited) counting statistics of aerosol particles. The major source of the random uncertainty of particle flux estimates is the non-stationarity of particle concentration as well as its flux (for flux random uncertainties see Fairall, 1984). The particle fluxes have typically large statistical uncertainty, in the order of 100% and more (Pryor et al., 2008, Rannik et al., 2003), therefore it is frequently difficult to determine whether the calculated upward particle occurrence reflects the true transport or was obtained by chance. Pryor et al. (2008)

investigated thoroughly the distribution and significance of upward fluxes as well as the relevance to several physical mechanisms causing them by taking into account also the error estimates of fluxes. They came to the conclusion of several possible physical mechanisms responsible for upward particle transport including the entrainment of particle-free air from above during the intensive ABL growth periods. Whitehead et al. (2010) observed similar systematic pattern over a tropical rain forest in case of supermicron particles. Upward particle fluxes were also observed on seasonal average diurnal patterns by Rannik et al. (2009a) in the statistical analysis of long-term particle flux measurements over a pine forest, confirming that the phenomenon is common over a long period of time.

Nilsson et al. (2001) also associated the occurrence of upward particle fluxes to the solar radiation increase and boundary layer development. In addition, they studied the evolution of the Aitken and Accumulation mode particle concentrations in the ML during the ABL growth and inferred the particle concentrations being entrained by using a simple ML growth model based on thermodynamical considerations. The model explained well the ML height as well as the particle concentration evolution. The entrained particle concentrations were determined to be virtually from 0% to 40% of the close-to-surface values, indicating that night-time horizontal advection was a dominating process at the site affecting the vertical profiles of aerosols above the SBL. The initialization of the aerosol concentration profiles during the first day of simulations in the current study represent such advective conditions and resulted in strong upward particle transport during the early morning ML growth. Whereas the night-time advection can be typical to SMEAR II site, it is certainly a site specific phenomenon and therefore for the rest of the period we intended to use the initialization of profiles with uniform particle concentration up to the residual layer height. Therefore our simulation results for the first day represent the conditions characteristic to strong horizontal advection and are during the rest of the days expected to underestimate the vertical transport due to ML growth.

Gordon et al. (2011) observed major fraction (60%) of upward particle fluxes for size interval 18 to 450 nm above a mixed forest in Ontario, Canada, by the EC technique. The upward particle flux rate was highest for 75 nm particles. One of the mechanisms for upward fluxes was the entrainment of clean air from aloft as discussed previously. As additional mechanism, the authors proposed the slowest growth rate of this particle size, suggesting that the authors referred to the aerosol dynamics as one of the reasons.

Pryor et al. (2013) also suggested the depletion mechanism as the most common cause of the upward fluxes above a sparse pine forest during the morning hours. Later in the day the authors attributed the upward fluxes of sub-30-nm particles to the growth of the newly formed particles by condensation of the BVOCs. All the mechanisms as the reasons for upward particles fluxes discussed here appear to be the plausible reasons according to our model simulations and can dominate depending on location, emission rates of BVOSs, time of day, particle size and possibly some other factors. The results of the current study identified the aerosol dynamics as one of the main mechanisms causing upward transport of particles with 30 nm in diameter and larger.

3.4.4 Fluxes of above 100 nm particles

~~We observed that~~Our results have shown that the aerosol and ABL dynamics can introduce significant systematic deviation of the exchange velocities above the canopy from dry deposition on the average. For around 100 nm particles the fluxes above the canopy exceeded the dry deposition sink and for larger than 100 nm the deposition was ~~very~~ poorly characterized by the fluxes above the canopy (see Table 1). The range of the flux to deposition ratio varied from negative to positive values, being especially large for about 100 nm particles, which coincides with the minimum of the particle deposition rate at this size. The median values presented in Table 1 were closer to unity than the averages. This implies that the averages are affected by extreme values corresponding to certain dynamical conditions occurring in the ABL. Such conditions certainly can take place in the real atmosphere. The fact that the median exchange velocities represent better deposition than the time average indicates that the median values are more robust statistics than the averages and should be perhaps used in representing the particle exchange instead of averages.

We note that the results based on model simulations were free of statistical uncertainty introduced by random errors to experimentally determined fluxes. Rannik et al. (2003) used a semi-empirical model to explain the size-integrated particle flux measurements performed at the same site with our model simulations. The model appeared to explain well the flux observation with particle population mainly consisting of below 100 nm particles. Deposition velocities for above 100 nm sizes were very uncertain. The authors proposed several reasons why the model was not able to explain the observations: presence of a mechanism controlling deposition of above 100 nm particles not described by the semi-empirical model as well as

several other reasons such as temporary pollution sources in the measurement source area. The possible reasons of meteorological origin were suggested to be horizontal advection of particle concentration, boundary layer growth and concentration dilution, and roll circulation in the ABL (e.g., Buzorius et al., 2001). This study has shown that such apparent uncertainty in deposition pattern of above 100 nm particles could be the case even in horizontally homogeneous conditions due to aerosol dynamical and ABL development processes.

4 Conclusions

Simulations performed by the model SOSAA coupling turbulent exchange within the ABL with detailed atmospheric chemistry and aerosol dynamics ~~We have observed~~indicated that the aerosol dynamics is strongly size-dependent but a significant source-sink term to aerosol concentration throughout the atmospheric column. Whereas the vertical transport is mostly compensating for particle loss inside the canopy due to the deposition, the aerosol dynamics leads to the concentration changes in the whole ABL. However, during the periods of intensive aerosol dynamics when new particle formation frequently occurs, the particle deposition and aerosol dynamics together with ABL development leads to complicated vertical transport patterns. For small particles (up to a few tens of nm) the deposition sink is relatively strong (compared to the aerosol dynamics) and the downward fluxes were predicted in the lower ABL. However, for some particle size ranges, depending on the aerosol dynamical processes, the stronger aerosol dynamical source inside and above forest (compared to higher ABL) can lead~~s~~ to upward particle transport such that the vertical fluxes above the canopy ~~are~~ might not be coherent with dry deposition ~~and therefore might do not represent the particle dry deposition~~ under such conditions. We have also observed that the ABL dynamics occasionally leads to upward particle transport which can be interpreted as the transport due to dilution of relatively particle-rich air within forest with the particle-poor air transported down from aloft during the active ABL growth phase.

The simulated turbulent transfer time scales inside the forest were much shorter than the time scales of deposition and aerosol dynamics for all sizes except the smallest at around 3 nm. In spite of efficient mixing inside the canopy, the particle fluxes at the canopy top ~~frequently~~ can deviate from the deposition rates inside forest. This is due to the transformation of aerosol concentration throughout the atmospheric column resulting in the complicated pattern of particle vertical transport. Therefore, the within-canopy deposition and transformation processes do not determine solely the particle vertical transport within and above the canopy

and the respective time scales are not sufficient to determine~~,~~ if the aerosol dynamics can cause significant particle flux divergence below the measurement level.

We conclude that under dynamical conditions studied here the ~~micrometeorological~~ particle fluxes ~~measurements conducted~~ above the forest canopy occasionally deviated ~~can be biased (at least in dynamical conditions studied here) in representing~~from the particle dry deposition sink inside the forest canopy. ~~The~~ Such deviations can be very large and for certain particle sizes ~~so that the conclusion applies also to the time averaged particle flux measurements~~even systematic after performing diurnal averaging of results.

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Appendix A. Description of measurements at SMEAR II

The SMEAR II (Station for Measuring Forest Ecosystem-Atmosphere Relations) field measurement station is located in Hyytiälä, Southern Finland (61° 51' N, 24°17' E, 181 m asl). The station is located in the area covered mainly by pine-dominated forests. The dominant height of the stand near the measurement tower was about 20 m in 2013. The main canopy at the site is characterized by the total leaf area index (LAI) ~6.5 $m^2 m^{-2}$ and stand density 1400 ha^{-1} (Launiainen et al., 2011). The forest floor vegetation is relatively low (mean height ~0.2–0.3 m) but dense (total LAI ~1.5 $m^2 m^{-2}$). However, in model setup a beta distribution of LAD was used that matched to observed turbulence statistics in and above the canopy and the forest floor vegetation as a separate layer was neglected (Boy et al., 2011). More detailed description of the station and the measurements can be found in Hari and Kulmala (2005).

Turbulent fluxes of momentum, heat, CO_2 and H_2O were measured by means of the EC technique. The system, located at 23 m height above the ground on the top of a scaffolding

tower, included an ultrasonic anemometer (Solent Research HS1199, Gill Ltd., Lymington, Hampshire, England) to measure the three wind velocity components and the sonic temperature, a closed-path infrared gas analyser (LI-6262, LiCor Inc., Lincoln, NE, USA) that measured the CO_2 and H_2O concentrations. The data were sampled at 21 Hz and a 2D rotation of sonic anemometer wind components and filtering to eliminate spikes were performed according to standard methods (e.g. Aubinet et al., 2000). The high-frequency flux attenuation was corrected by using empirical transfer functions and co-spectral transfer characteristics (Mammarella et al., 2009).

Aerosol size distribution (from 3 nm to1 µm) measurements were performed using a Differential Mobility Particle Sizer (DMPS) system. The aerosol was sampled from inside the forest at 2 m height. Details of the DMPS measurement system are presented in Aalto et al. (2001).

Appendix B. Initialisation of model SOSAA

The chemistry scheme employed by the model for this study included the relevant Master Chemical Mechanism (MCM) chemical paths (Jenkins et al., 1997; Jenkins et al., 2003; Saunders et al., 2003) for the following parent molecules: methane, methanol, formaldehyde, acetone, acetaldehyde, MBO, isoprene, alpha-pinene, beta-pinene, limonene and beta-caryophyllene. For other emitted organic compounds including myrcene, sabinene, 3-carene, ocimene, cineole and "other" monoterpenes, and farnesene and "other" sesquiterpenes, the MCM chemistry paths are not available and we thus included their first-order oxidations with OH, O_3 and NO_3. For the reactions of the stabilized Criegee intermediates (sCI) from alpha- and beta-pinene and limonene, we used the rates from Mauldin III et al. (2012), similar to ñScenario Cò in Boy et al. (2013). For the sCI from isoprene, we used the rates from Welz et al. (2012) as done in ñScenario Dò in Boy et al. (2013). Sulfuric acid and nitric acid were removed from the gas phase based on the condensation sinks calculated from background aerosol loading.

There was no specific initialization of chemistry state for the model (all variables were initialized as zero while created). Exceptions were the passive tracer concentrations (CO_2, NO, NO_2, SO_2 and O_3), which were initialized with measurements. The concentrations of these five passive tracers were always read in from measurements during the simulation. The

time resolution of input data was half an hour and the data was linearly interpolated for each time step in model run. The vertical profiles of the particle concentrations were initialized each night as described in Sect. 2.2.

Global short wave radiation, top boundary temperature, humidity and wind speed were fixed to inputs throughout simulation. The global short wave radiation was measured at SMEAR II. Temperature, humidity and wind speed at the top boundary were based on ECMWF reanalysis data.

The initial temperature profile was assumed linear, using the input top border temperature and input temperature gradient. The wind profile was set using the logarithmic wind law, the roughness and wind speed at the top boundary. Initial humidity was taken constant throughout the ABL and the heat fluxes and TKE were set to zero. Mixing length was initialized as $l = 0.40\frac{z + z_0}{1 + 0.016z}$.

At the lower boundary, soil humidity for the uppermost layer was set to 0.2 kg kg^{-1}. Soil temperature was set -2 degrees from air temperature at the lowest level and leaf temperature was set equal to air temperature. Heat flux to the soil was based on the measurements from the SMEAR II station throughout simulations.

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Table 1. Statistics of the ratio of the flux at the canopy top to deposition sink integrated over the canopy over 10 days period in May 2013. The average statistics $\langle V_e \rangle$ and $\langle |V_{dep}| \rangle$ were averaged over the simulation period first and then the ratio was found, whereas the percentile statistics apply for the ratios $\frac{V_e}{|V_{dep}|}$ obtained from model simulations for each 10 minute period.

Particle size (nm)	**3**	**10**	**30**	**100**	**300**	**850**	**3-30**	**30-100**	**100-1000**
$\frac{\langle V_e \rangle}{\langle \lvert V_{dep} \rvert \rangle}$	0.90	0.99	1.36	2.09	0.53	0.82	1.11	1.99	0.66
Q5	-0.24	-0.18	-0.32	0.15	-1.30	0.20	0.33	0.70	-0.76
Q25	0.77	0.87	0.93	0.82	0.32	0.73	0.97	0.95	0.56
Median	0.97	1.00	1.06	0.94	0.85	0.92	1.06	1.04	0.86
Q75	1.15	1.09	1.31	1.34	0.92	0.96	1.20	1.57	0.92
Q95	1.81	1.70	3.36	9.59	1.01	1.00	2.12	10.5	0.98

Table 2. Statistics of the ratio of the flux at the canopy top to deposition sink integrated over the canopy over 10 days period in May 2013. For more details see Table 1. Morning refers to time period from sunrise till noon, afternoon from noon till sunset and night from sunset till sunrise.

Time	**Morning**			**Afternoon**			**Night**		
Particle size (nm)	3-30	30-100	100-1000	3-30	30-100	100-1000	3-30	30-100	100-1000
$\frac{\langle V_e \rangle}{\langle \lvert V_{dep} \rvert \rangle}$	1.25	2.92	0.48	1.12	1.67	0.73	0.84	1.19	0.81
Q5	0.68	0.20	-1.36	0.77	0.70	0.07	-0.27	0.87	-0.16
Q25	1.04	0.94	0.33	0.97	0.94	0.56	0.92	0.96	0.77
Median	1.17	1.17	0.84	1.03	1.03	0.84	1.03	1.01	0.86
Q75	1.46	2.96	0.91	1.13	1.40	0.91	1.11	1.17	0.93
Q95	2.21	29.6	0.98	1.69	7.21	0.99	3.85	3.04	0.96

Figure captions

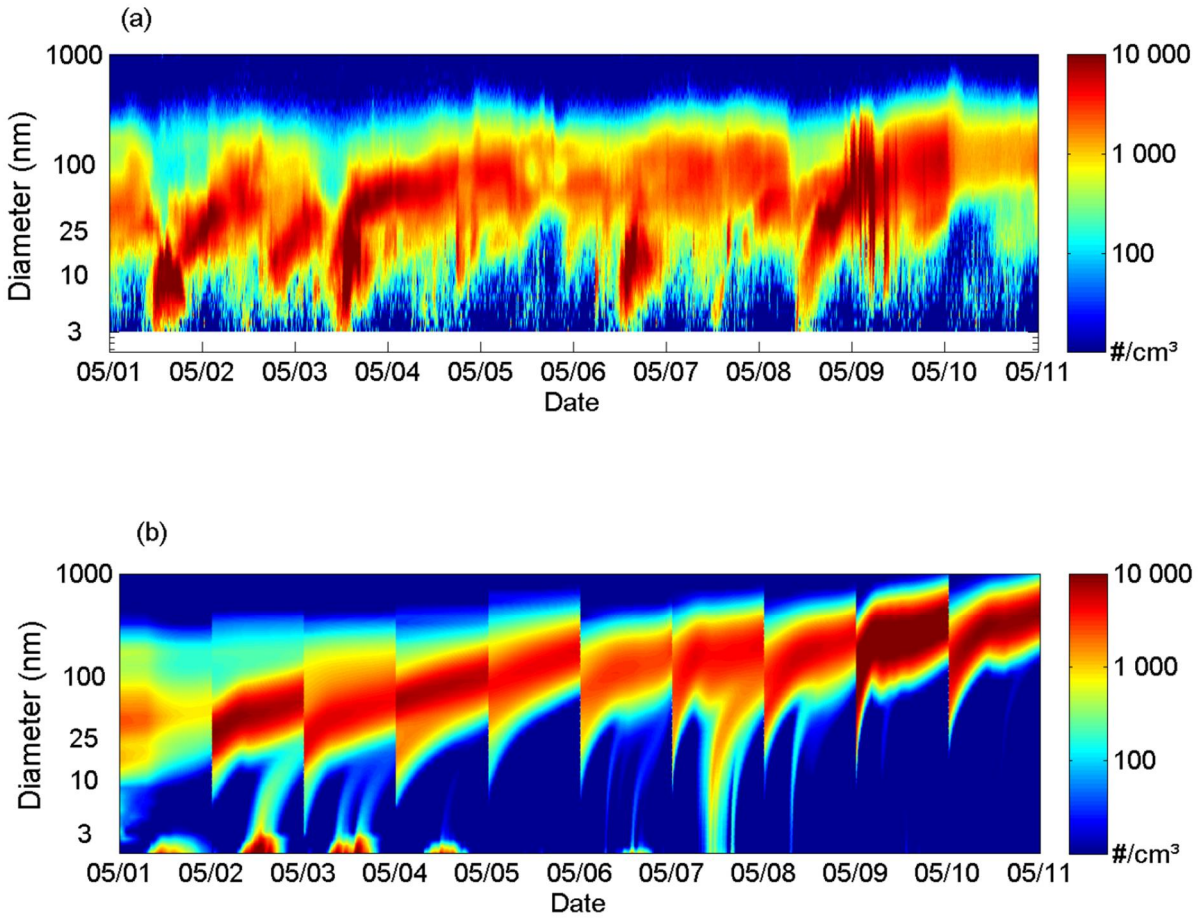


Figure 1. Aerosol size distribution at 2 m height during 10 days period in May 2013 as (a) measured by the DMPS system and (b) predicted by the model SOSAA.

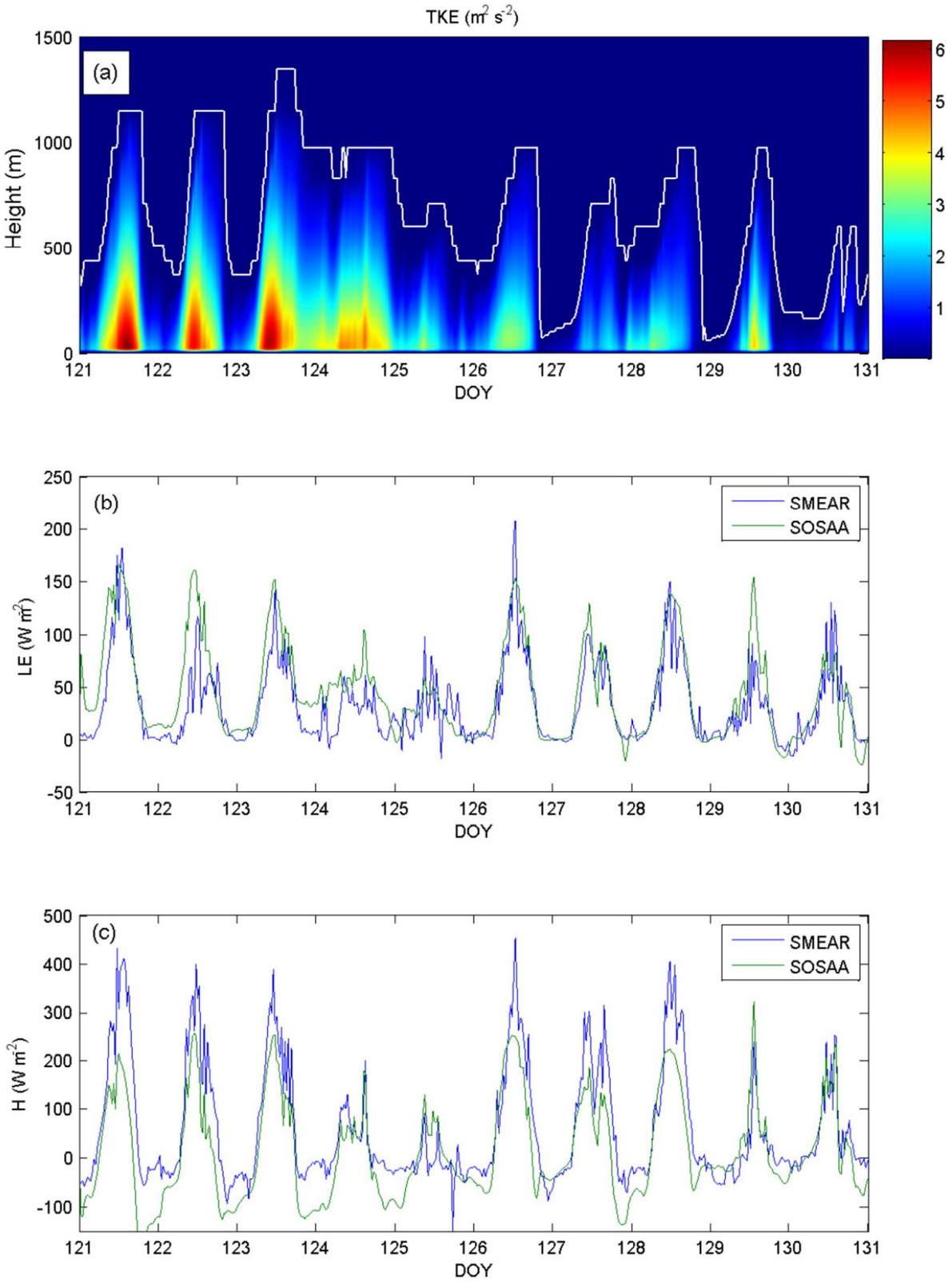


Figure 2. General meteorology: (a) TKE and ABL height, (b) latent heat flux LE and (c) sensible heat flux H during 10 days period in May 2013. SMEAR refers to measurements at the station.

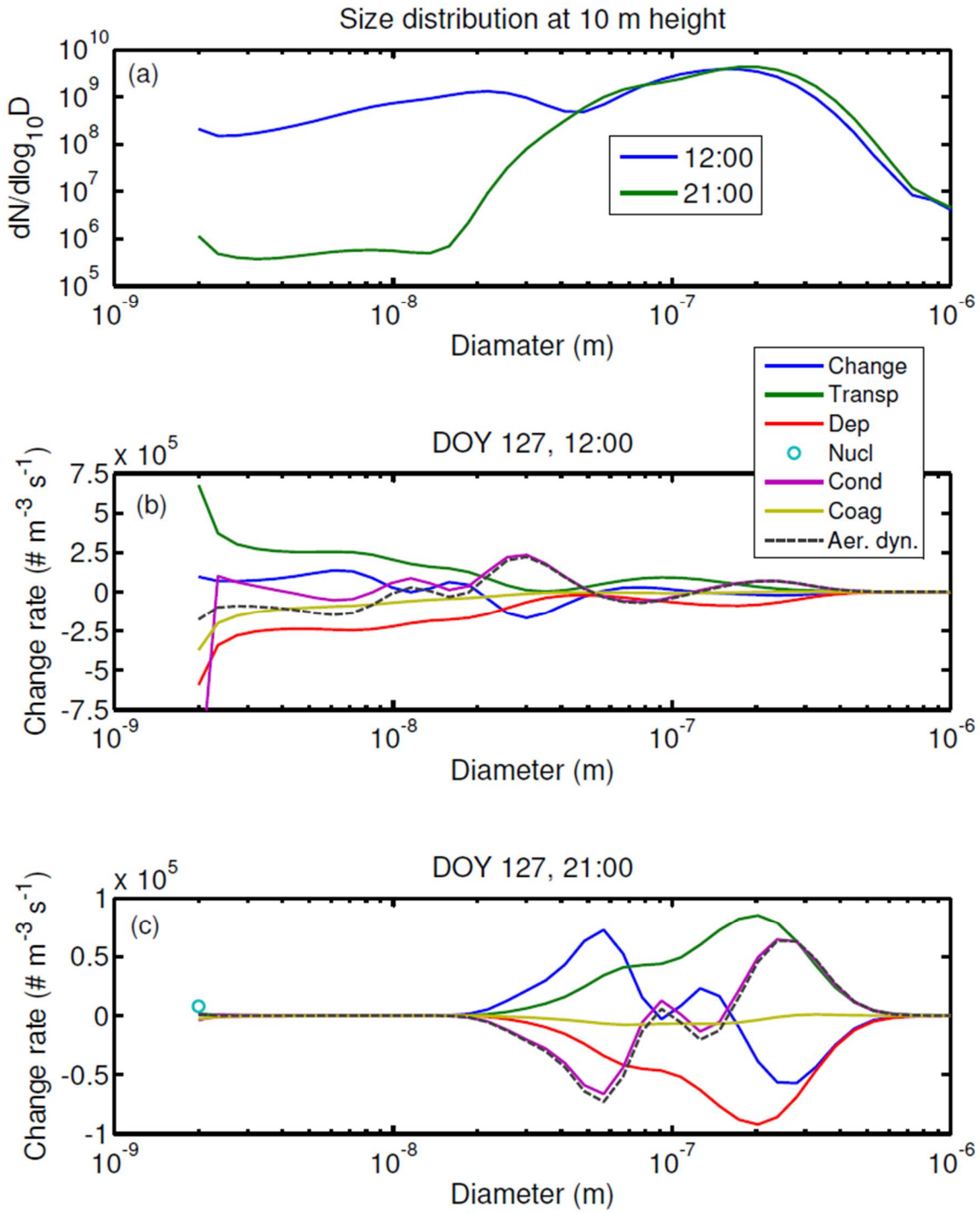


Figure 3. (a) Aerosol size distributions and the conservation terms at (b) 12:00 LT (the values for nucleation and condensation terms at 2 nm are out of scale, being in absolute values about 1.3x10^6 # m^3 s^{-1} but opposite in sign) and (c) 21:00 LT as a function of particle size at 10 m height 07 May. The storage change (Change), the (vertical) transport (Transp), the particle deposition (Dep) and the aerosol dynamical (Aer. Dyn.) terms denote the respective terms in Eq. (4). The aerosol dynamical term is the sum of the numcleation (Nucl), condensation growth (Cond) and coagulation (Coag) terms.

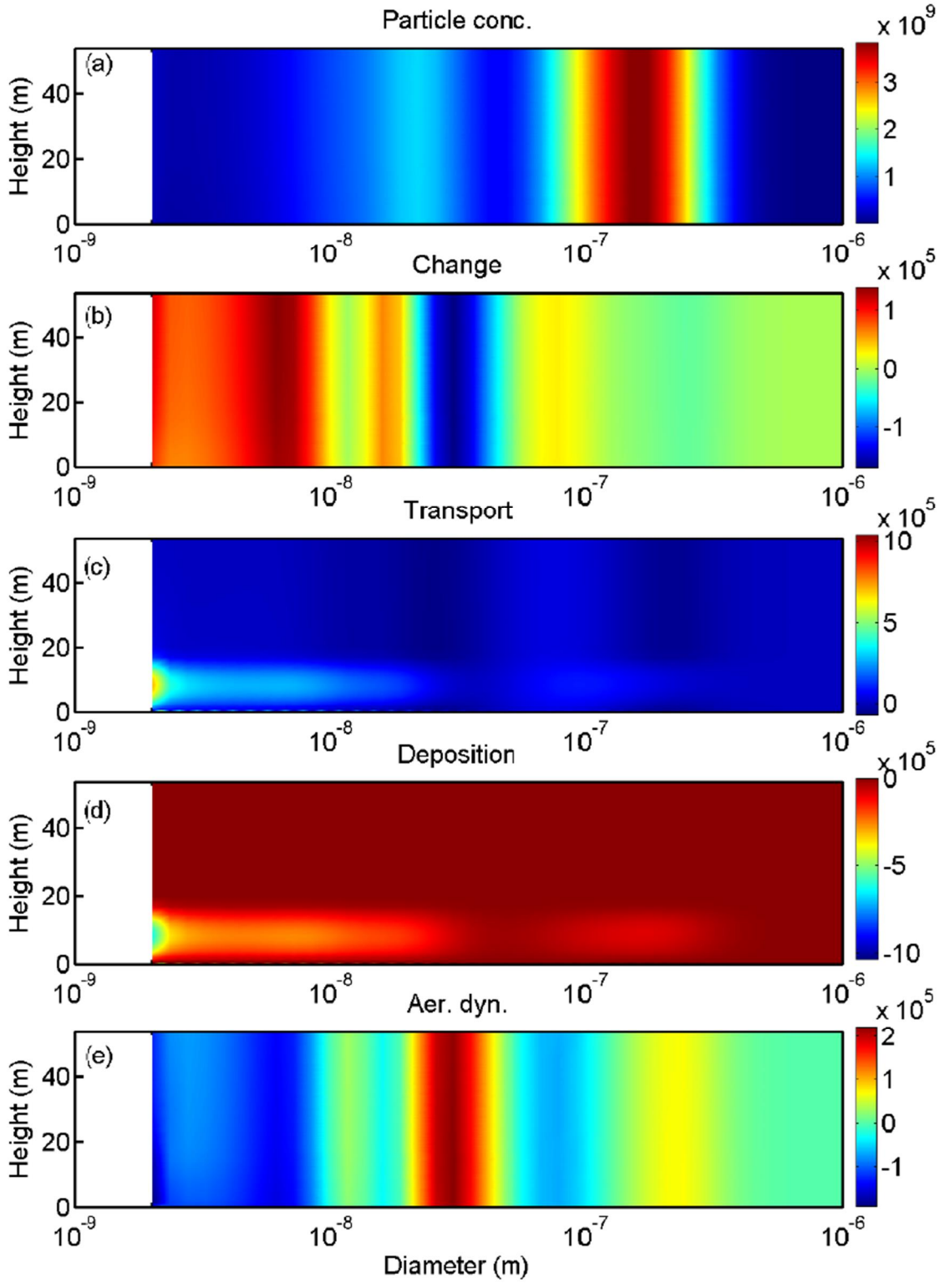


Figure 4. Vertical profiles of aerosol (a) number concentration (# m^{-3}) and conservation terms: (b) storage change (# m^{-3} s^{-1}), (c) transport (in # m^{-3} s^{-1}), (d) deposition (in # m^{-3} s^{-1}), (e) aerosol dynamical (in # m^{-3} s^{-1}) on 07 May at 12:00 LT for particle size range from 2 nm to 1 μm.

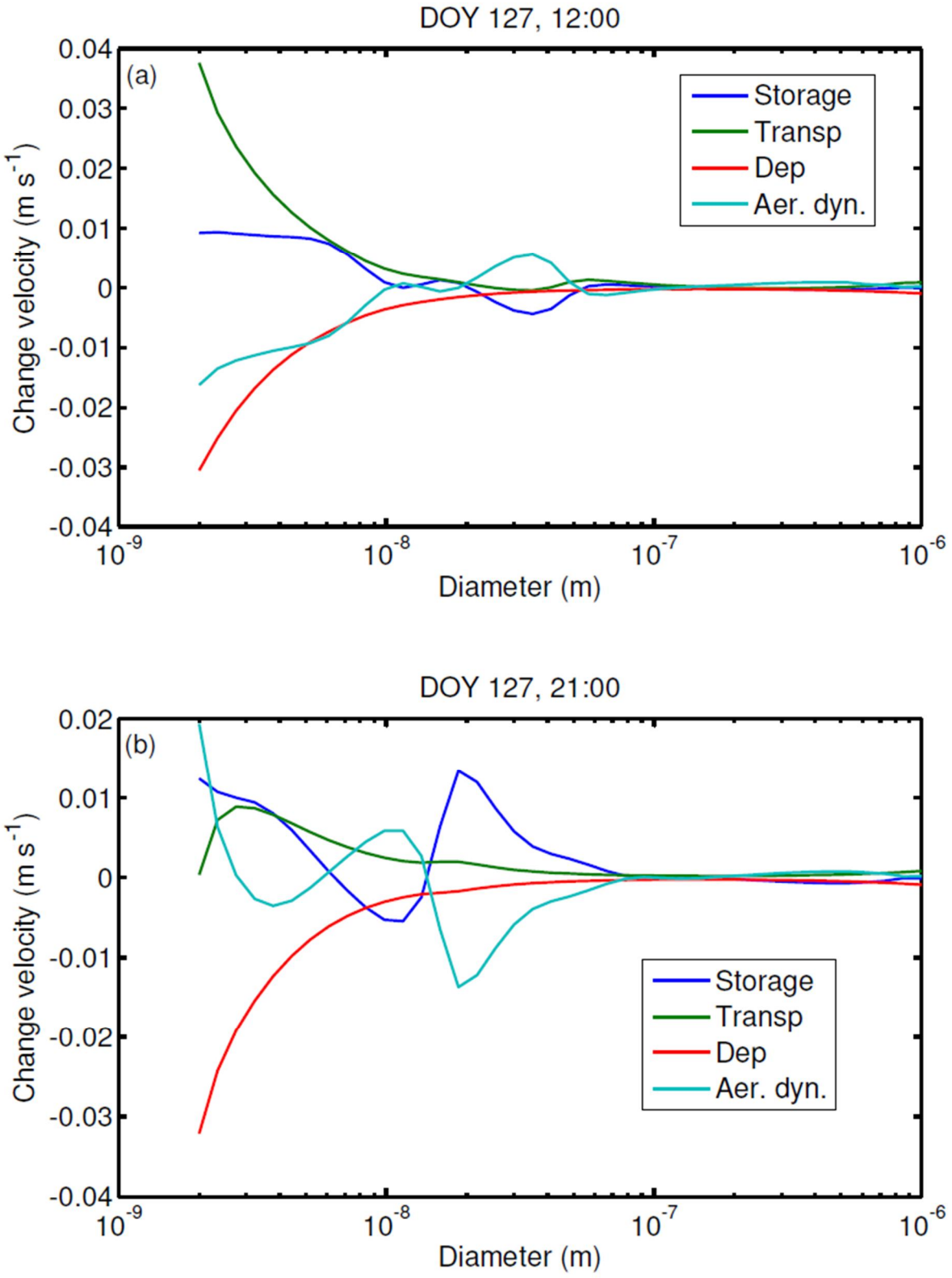
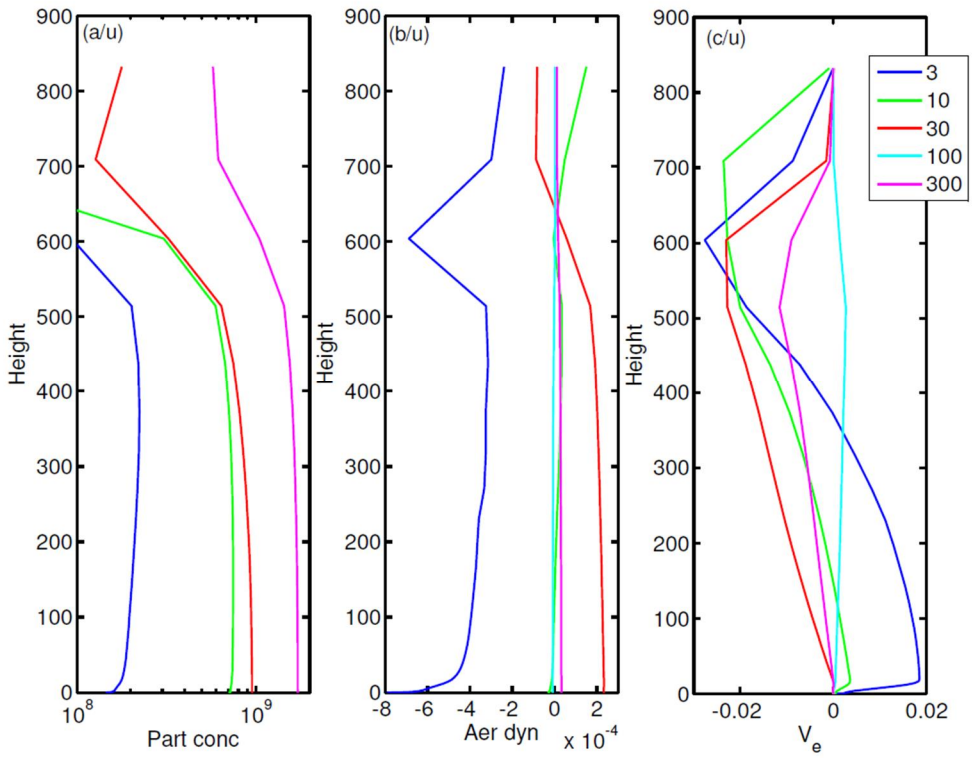


Figure 5. Integrated up to the canopy top conservation equation (Eq. 4) terms for the same periods as in Fig. 3 (a) and (b), normalised with the concentration at the canopy top.



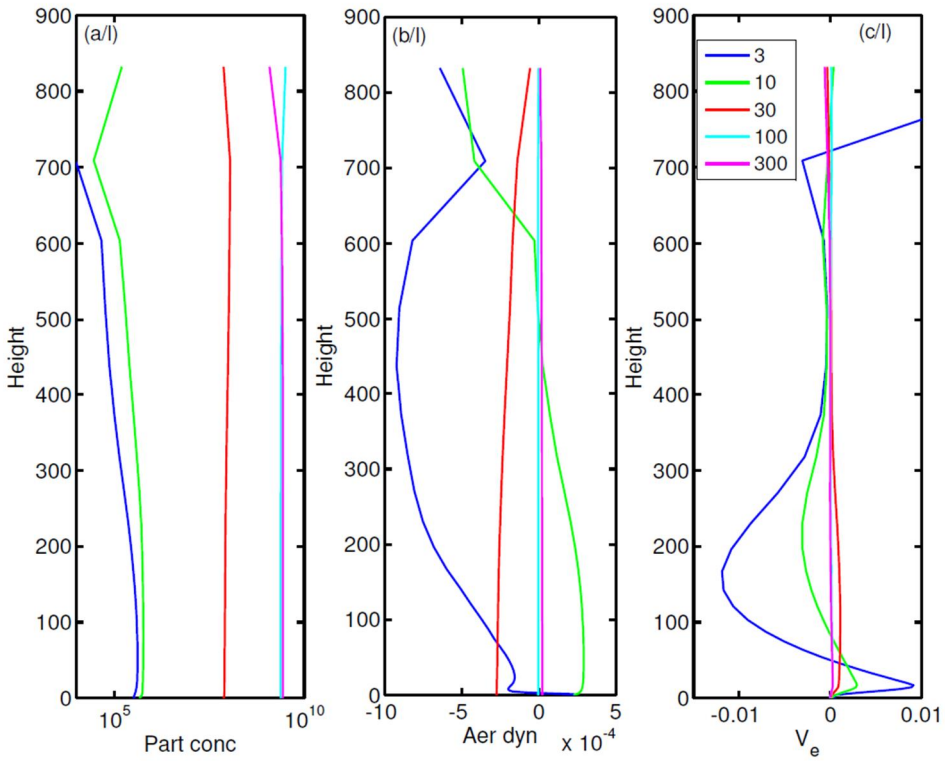


Figure 6. Vertical profiles of (a) the particle concentration (# m^{-3}), (b) change rate due to aerosol dynamics (s^{-1}), and (c) the vertical exchange velocity defined to be positive for downward transport (m s^{-1}) for selected particle sizes 07 May at 12:00 LT (upper panels denoted by /u) and 21:00 LT (lower panels denoted by /l). For panels (b) and (c) normalisation with local concentrations was used.

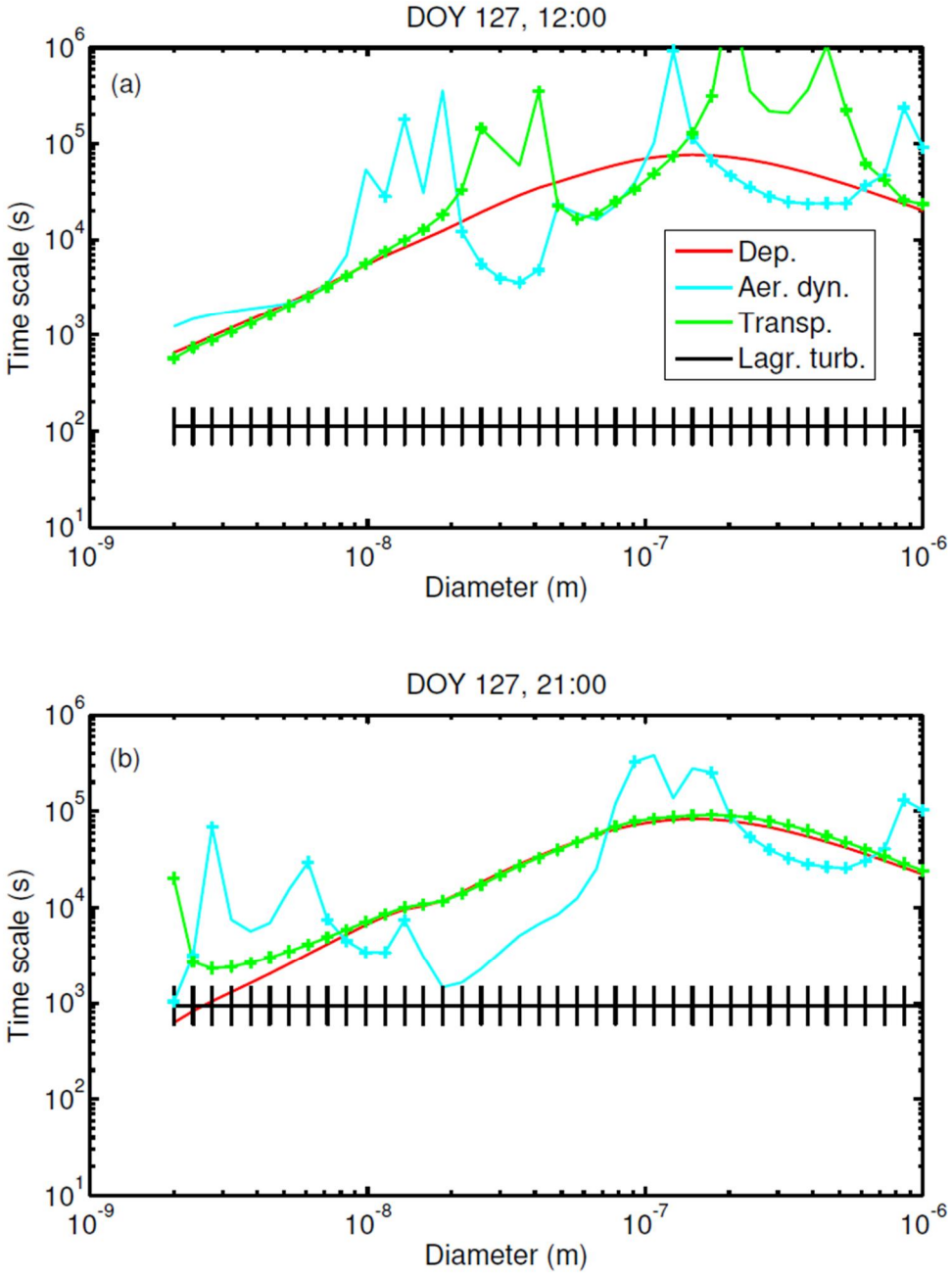


Figure 7. The time scales of deposition, aerosol dynamics and transport (equivalent to vertical exchange) as defined by Eqs. (9) together with (5), (6) and (7) at (a) 12:00 LT (the values for the transport term are out of scale at about 200 and 450 nm, being about $+3.7 \times 10^6$ and -1.05×10^6 s, respectively) and (b) 21:00 LT 07 May 2013. In addition the Lagrangian time scale for turbulent transfer (corresponding to aerodynamic resistance only) as simulated according to Eq. (10), being presented as the median air parcel travel time between the forest floor and the canopy top with upper and lower quartiles. The „+“ sign reflects the positive sign of the respective term (the source), whereas no such sign infers the negative (sink) term.

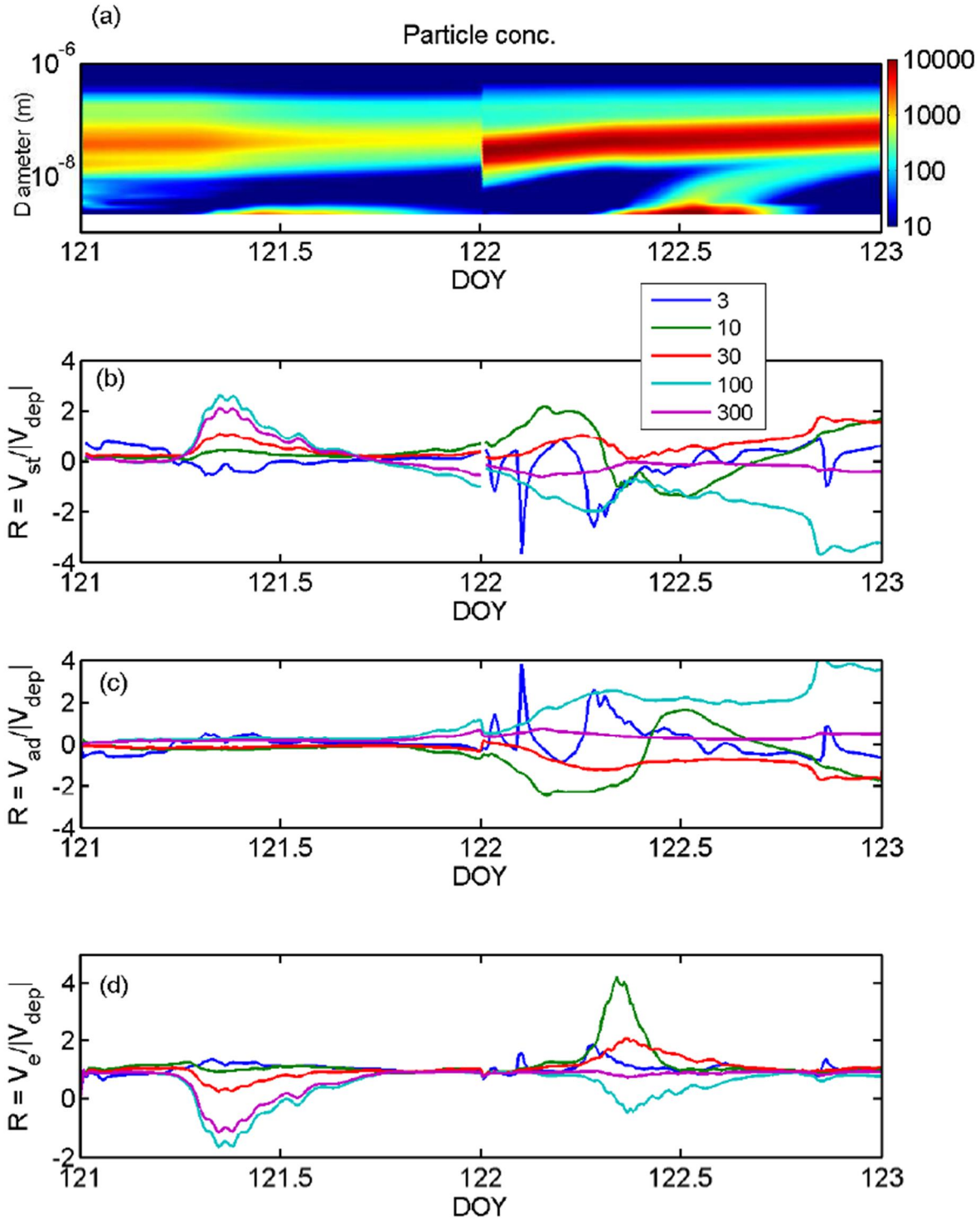
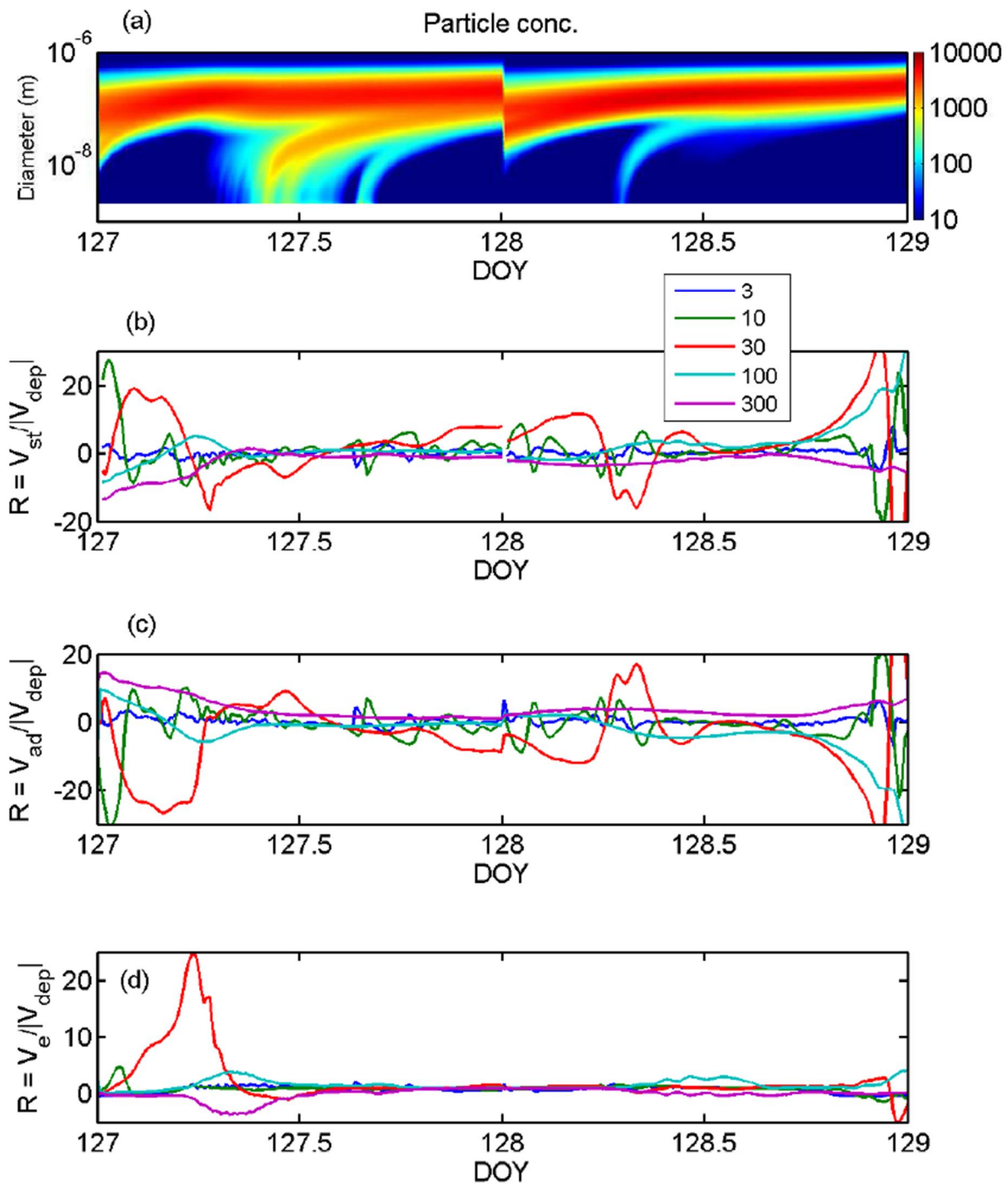


Figure 8. (a) Particle size spectrum, and the change velocities (presented as the ratios to the absolute value of the deposition term) for selected particle sizes for (b) storage, (c) aerosol dynamics and (d) vertical exchange during 01 and 02 May (DOY 121 and 122) 2013.



~~Figure 9~~. ~~(a) Particle size spectrum, and the change velocities (presented as the ratios to the absolute value of the deposition term) for selected particle sizes for (b) storage, (c) aerosol dynamics and (d) vertical exchange during May 7th and 8th (DOY 127 and 128) 2013.~~

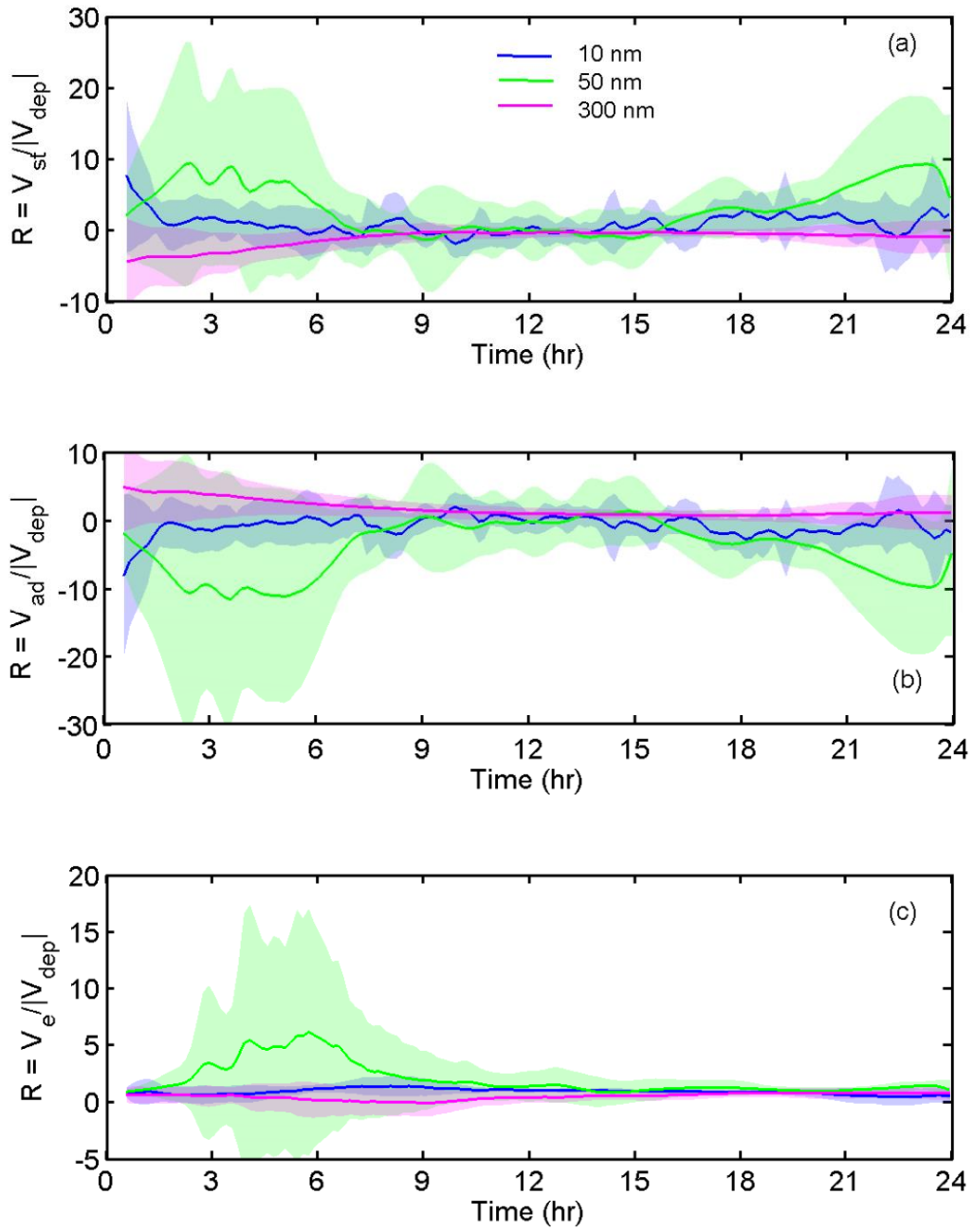


Figure 9. Diurnal variation of change velocity for (a) storage, (b) aerosol dynamics and (c) vertical exchange for selected particle sizes. The lines present the ratios of the average change velocities to the average deposition term according to $\frac{\langle V \rangle}{\langle |V_{dep}| \rangle}$ obtained from model simulations for each 10 minute period and the shaded areas the variation range as $\pm \frac{\sigma_V}{\langle |V_{dep}| \rangle}$ around the averages.

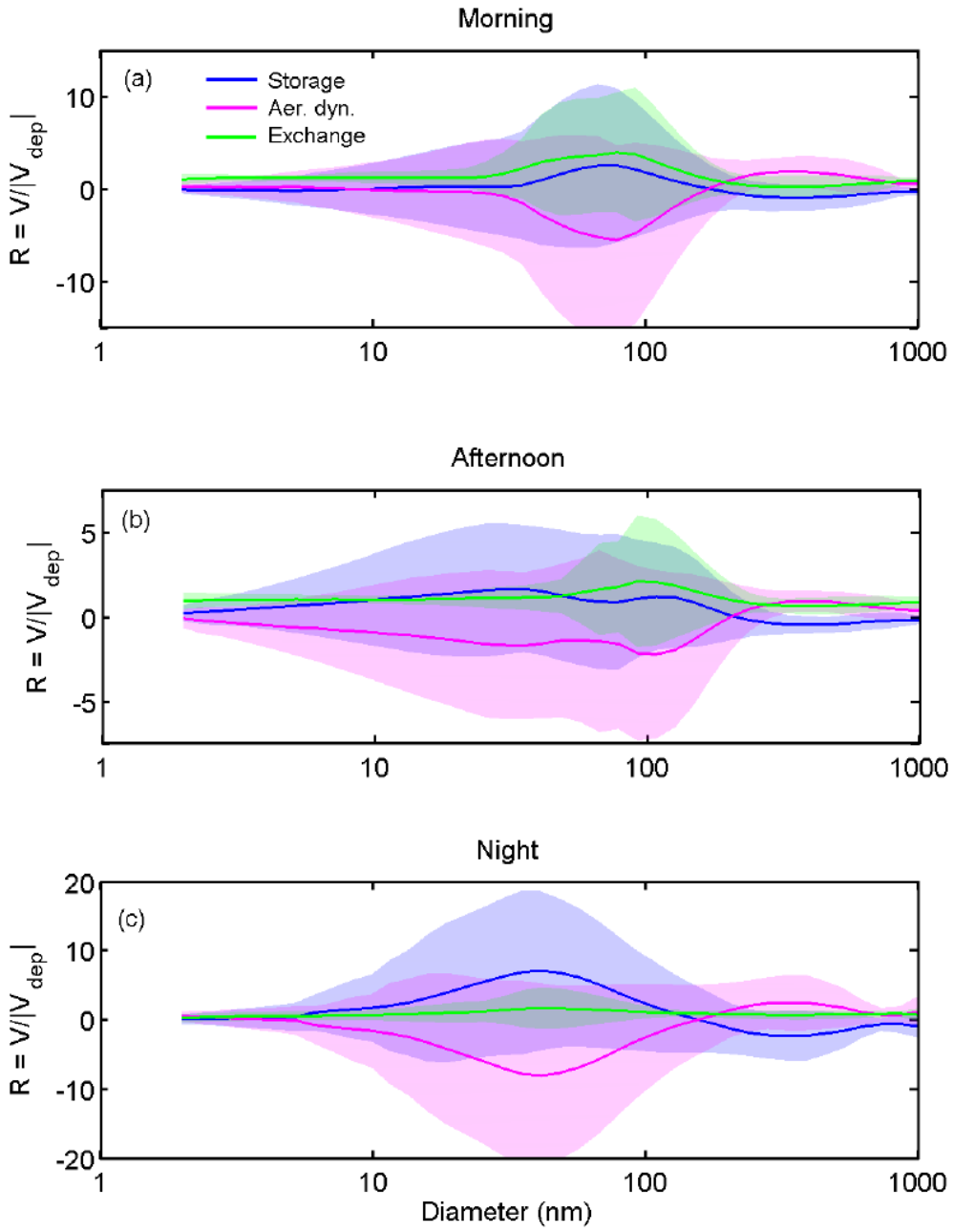


Figure 10. Variation of change velocities with particle size for (a) morning (from sunrise till noo), (b) afternoon (from noon till sunset) and (c) night (sun below horizon) periods for: blue line the storage, magenta the aerosol dynamics and green the vertical exchange velocities. The lines present the ratios of the average exchange velocities to the average deposition term according to $\frac{\langle V_e \rangle}{\langle |V_{dep}| \rangle}$ obtained from model simulations for each 10 minute period and the shaded areas the variation range as $\pm \frac{\sigma_{V_e}}{\langle |V_{dep}| \rangle}$ around the averages.

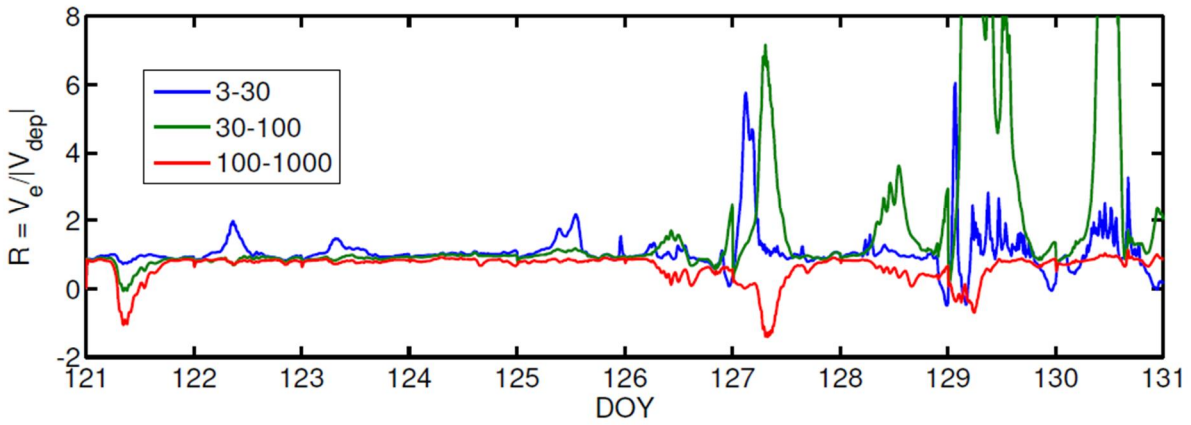


Figure ~~10~~11. The exchange velocity V_e at the canopy top for selected particle size intervals during 10 days period in May 2013 normalised with the absolute value of the deposition velocity $|V_{dep}|$. Peak values for the size range 30-100 nm at doy 129 and 130 were about 30-35.