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SOSA – a new model to simulate the concentrations of organic vapours and sulphuric acid inside the ABL – Part 1: Model description and initial evaluation

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Abstract

Chemistry in the atmospheric boundary layer (ABL) is controlled by complex processes of surface fluxes, flow, turbulent transport, and chemical reactions. We present a new model SOSA (model to simulate the concentration of organic vapours and sulphuric acid) and attempt to reconstruct the emissions, transport and chemistry in the ABL in and above a vegetation canopy using tower measurements from the SMEAR II at Hyytiälä, Finland and available soundings data from neighbouring meteorological stations. Using the sounding data for upper boundary condition and nudging the model to tower measurements in the surface layer we were able to get a reasonable description of turbulence and other quantities through the ABL. As a first application of the model, we present vertical profiles of organic compounds and discuss their relation to newly formed particles.

1 Introduction

While modern measurement techniques can characterize turbulent flow and transfer in the surface layer, the data does not serve for all of our needs for understanding the processes in the vegetation-atmosphere interface. Vertical profiles (e.g. meteorological soundings) of the boundary layer are limited to specific points in time, and time series data are collected only on some levels, usually by instruments mounted on a tower. Chemical reactions in the atmosphere involve many different compounds, originating from different source locations. To properly describe the complex chemistry in the surface and boundary layer, we need to know the transport rates of the molecules of interest with good accuracy. One way to reconstruct the dynamical and spatial structure of flow, turbulence and transport of quantities in the soil-vegetation-atmosphere system is numerical modelling. Modelling provides complete information on all variables of interest, including quantities on which measurements are scarce, or which are not measured at all.

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Among the most interesting and important chemical compounds in this sense are the products of highly reactive organic molecules like sesquiterpenes. The lifetime of many sesquiterpenes, with regard to reaction with ozone, is of the order of minutes and only a small fraction of these very volatile organics can reach the upper part of the atmospheric boundary layer (ABL), depending on the strength of turbulent mixing. These molecules and their reaction products are of interest as they potentially play a major role in atmospheric nucleation as pointed out by Bonn et al. (2008 and 2009). A better understanding of the mixing and the vertical distribution seems to be crucial for improving our knowledge concerning the part of the troposphere where new particles are formed.

The aim of this paper is to demonstrate a modelling approach as a tool for the description of the transport of biochemical compounds in the vegetation-atmosphere system and to be used for investigation of complex processes like the formation of secondary organic aerosols (SOA). Thus, the paper describes a model with verification, intended to be a tool for further applications in environmental research. To test the model we use observations from Hyytiälä, Southern Finland.

2 Measurements

Most of the measurements, used as input data and for verification, are from the Station to Measure Ecosystem-Atmosphere Relation (SMEAR II) in Hyytiälä, Finland. A detailed description of the station and instrumentation is given by Kulmala et al. (2001) and in http://www.atm.helsinki.fi/SMEAR/. For lower model boundary conditions, meteorological measurements (temperature, humidity, wind velocity) at several levels inside and above the canopy, between surface and 72 m height, were utilized for nudging to improve the model results. Spectral irradiance measurements from the station (Boy et al., 2002) were used as inputs for photochemical reaction rates. To calculate the condensation sinks for sulphuric and nitric acid we used particle size distributions at the surface based on DMPS (Differential Mobility Particle Sizer) and APS (Aerodynamic Particle Sizer) measurements from SMEAR II.

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The model was evaluated by comparing modelled temperature, friction velocity, turbulent sensible and latent heat fluxes with observations. The observed fluxes were based on eddy covariance measurements calculated as 30 min block-averaged covariance. Wind components and temperature were measured with an ultrasonic anemometer (Solent HS1199) and water vapour with a high frequency gas analyser (Li-Cor 6262, infrared absorption) at 23 m height. Friction velocity u^* describes the strength of mixing (momentum flux) and is calculated from the covariance of horizontal and vertical wind fluctuations. Similarly, observed sensible and latent heat fluxes are based on the covariance of measured vertical wind velocity and temperature and humidity fluctuations, respectively. Mammarella et al. (2009) have discussed the uncertainties of measurements and concluded that the aging (contamination) of sample lines may lead to 10–15% underestimation in humidity flux during summer time.

Meteorological conditions at the upper boundary of the model were determined by utilizing soundings which provided information on temperature, humidity and wind conditions. Jokioinen sounding station is located ∼100 km south from Hyytiälä and Tikkakoski station ∼100 km north-east from Hyytiälä. The soundings were done at 06:00 and 18:00 UTC in Tikkakoski and at 00:00 and 12:00 UTC in Jokioinen. Input values of variables at the upper border of the model were interpolated linearly between observation times.

3 Model approach

SOSA is a combination of (i) a meteorological boundary layer model, also describing the biophysical conditions inside a plant canopy in a fine resolution, (ii) a physiological model of plant canopy BVOC emissions, and (iii) chemical kinetics model for a large amount of chemicals and reactions occurring at different levels of the atmosphere model.

Reactive chemicals originate from the vegetation according to the BVOC emission model, the meteorology model describes the environmental conditions (such as

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temperature, humidity, solar radiation in different wavelengths) affecting the chemical reaction rates inside the vegetation canopy and in the atmosphere. The chemical kinetics model describes the time evolution of the concentrations of the chemical species as they react with each other. The meteorology model also describes the mixing and transport of chemicals between the layers of atmosphere in the model.

SOSA is implemented in Fortran, and the chemistry module utilizes parallel computing, so when running on a computer cluster, the chemistry in different atmospheric layers can be computed in parallel.

3.1 Transport model

The meteorological transport model is based on the coupled plant-atmosphere boundary layer model SCADIS (Sogachev et al., 2002, 2005; Sogachev and Panferov, 2006). For canopy flow, the model was extensively tested against field and wind tunnel experiments by Sogachev and Panferov (2006) and demonstrated reasonable performance and universality. Numerical tests showed also the practical applicability of the approach used for the description of buoyancy effect. For example, the model properly reproduced both the surface layer wind, as estimated from the Monin-Obukhov similarity theory, and the mixing height evolution, as observed above forested terrain in Southern Finland (Sogachev, 2009). The scalar transport description was also tested by Sogachev et al. (2002, 2008).

SCADIS is based on the Reynolds averaged Navier-Stokes equations for turbulent flow (Pope, 2000). Turbulent fluxes are expressed as the product of a turbulent diffusion coefficient and the gradient of a mean quantity according to the concept first proposed by Boussinesq (e.g. Pielke, 2002). Concentrating on vertical exchange we use a one dimensional version of the model, in which the evolution equations for momentum, heat and moisture are:

$$\frac{\partial u}{\partial t} = f(v - v_g) + \frac{\partial}{\partial z} K \frac{\partial u}{\partial z} + S_u \quad (1)$$

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$$\frac{\partial v}{\partial t} = -f(u - u_g) + \frac{\partial}{\partial z} K \frac{\partial v}{\partial z} + S_v \tag{2}$$

$$\frac{\partial T}{\partial t} = \frac{\partial}{\partial z}\left[\frac{K}{\sigma_H}\left(\frac{\partial T}{\partial z} + \gamma_a\right)\right] + R + S_T \tag{3}$$

$$\frac{\partial q}{\partial t} = \frac{\partial}{\partial z}\frac{K}{\sigma_V}\frac{\partial q}{\partial z} + S_q. \tag{4}$$

Here, z is the local vertical axis, t represents time, u and v are the velocity components along the east and the north axes, respectively, u_g and v_g are the components of geostrophic wind (over the atmospheric boundary layer), f is the Coriolis parameter, K is the kinematic eddy viscosity (or turbulence coefficient), T is air temperature and q is air specific humidity. S notes the source/sink associated with the vegetation effect for the corresponding variable indicated by subscript (see below for an expression). In Eq. (3) R describes the long wave cooling rate, γ_a is the dry adiabatic lapse rate ($\gamma_a = 0.0098\,\mathrm{K\,m^{-1}}$), σ_H and σ_V are the Prandtl number and the Schmidt number for heat and water vapour, respectively. They are assumed to be equal to each other, i.e. $\sigma = \sigma_H = \sigma_V$, and approximated by the following empirical formulations (Businger et al., 1971; Sogachev et al., 2002):

$$\sigma^{-1} = \begin{cases} 1.35(1 + 1.35\,Ri)^{-1} & \text{for } Ri \geq 0 \\ 1.35(1 - 15\,Ri)^{1/4} & \text{for } Ri < 0 \end{cases} \tag{5}$$

where Ri is the local gradient Richardson number. The model is based on the $E - \omega$ closure scheme (E is the turbulent kinetic energy (TKE) and $\omega = \varepsilon/E$ is the specific dissipation, where ε is the dissipation rate of TKE) and includes two evolution equations for these variables (Wilcox, 2002):

$$\frac{\partial E}{\partial t} = \frac{\partial}{\partial z}\left(\frac{K}{\sigma_E}\frac{\partial E}{\partial z}\right) + P + B - \omega E + S_E \tag{6}$$

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$$\frac{\partial \omega}{\partial t} = \frac{\partial}{\partial z}\left(\frac{K}{\sigma_\omega}\frac{\partial \omega}{\partial z}\right) + \frac{\omega}{E}\left(C_{\omega 1}P - (C^{-}_{\omega 2}C_{\omega 1})B\right) - C_{\omega 2}\omega^2 + S_\omega \quad (7)$$

Equations (6) and (7) are modified for description of canopy and buoyancy effects (Sogachev and Panferov, 2006; Sogachev, 2009) as follows.

$$P = K\left(\left(\partial u/\partial z\right)^2 + \left(\partial v/\partial z\right)^2\right) \quad (8)$$

is the production term of TKE due to wind shear, and

$$B = -\left(K/\sigma\right)\left[\left(g/T\right)\left(\partial T/\partial z + \gamma_a\right) + 0.618\left(g/\rho\right)\left(\partial q/\partial z\right)\right] \quad (9)$$

is the production term of TKE due to buoyancy. Here g is the gravitational acceleration and ρ is reference air density and σ_E and σ_ω are the Schmidt numbers for E and for ω, respectively. K is defined as

$$K = C_\mu E^{1/2}/\omega \quad (10)$$

where the coefficient C_μ (squared ratio of equilibrium shear stress to TKE) is the key parameter for two-equation schemes, and whose estimates vary considerably from experiment to experiment: typical values in the ABL vary within the range 0.03–0.12. The full information and discussion about how original constants ($\sigma_E = \sigma_\omega = 2$, $C_{\omega 1} = 0.52$ and $C_{\omega 2} = 0.833$) derived from wind tunnel studies (Wilcox, 2002) can be adapted for ABL simulation is presented by Sogachev and Panferov (2006).

Source/sink terms describing vegetation-air interactions are:

$$S_{\varphi=u,v} = -c_d A U \varphi \quad (11)$$

$$S_{\varphi=T,q} = A_\varphi\left[\eta\, g^{sn}_\varphi\left(\varphi^{sn}_l - \varphi\right) + (1-\eta) g^{sd}_\varphi\left(\varphi^{sd}_l - \varphi\right)\right] \quad (12)$$

$$S_E = 0 \quad (13)$$

$$S_\omega = \left(C^{-}_{\omega 2}C_{\omega 1}\right) 12 C_\mu^{1/2} c_d A U \omega \quad (14)$$

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Here U is the mean wind speed, c_d is the drag coefficient including the shelter effect, and $A(z)$ is the projected leaf area per unit volume (leaf area density – LAD). Leaf area density is related to the leaf area index (LAI) by

$$\mathrm{LAI} = \int_0^h A(z)\,dz \tag{15}$$

where h is the canopy height. A_ϕ defines the total leaf surface area density taking part in scalar exchange with the surrounding air (= $2A$ for leaves and = $2.7A$ for needles). η is the sunlit leaf surface area fraction, ϕ is the atmospheric value of the scalar and φ_l^{sn} and φ_l^{sd} are the scalar values on the leaf surface for sunlit and shaded fraction, respectively. Integral coefficients for the scalar exchange between the phytoelements and canopy air (g_φ^{sn}, g_φ^{sd}) are calculated taking into account the stomatal conductance to scalar transfer and the relevant aerodynamic resistance. We refer to Sogachev et al. (2002) for details about the full equations and parameterizations used for the estimation of source terms in Eq. (12) and boundary conditions for T and q.

The transport of any other passive tracer C is described in the model by an equation analogous to Eq. (4) with a source term S_c analogous to Eq. (12) and corresponding boundary conditions.

The system of nonlinear differential equations of turbulent flow inside and above the non-uniform vegetation is integrated numerically with boundary conditions imposed at the soil surface and the top of atmospheric boundary layer. The tridiagonal matrix algorithm (TDMA) was utilized to solve the system of algebraic equations arising from the discretization of the transport equations. For the discretization of the computational domain, a cell-centered finite volume approach was used (Patankar, 1980). A vertical grid with 75 height levels was used, with logarithmically increasing step size from soil surface up to 3000 m height. The smallest step was 17 cm at the surface, increasing to 200 m at the upper boundary. An implicit scheme for time integration in the model allows using an arbitrary time step, however to avoid loss of information important for passive transport, a time step of 10 s was used.

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In the nudging scheme an extra tendency term is added to each prognostic equation which forces the predicted variable towards the available observations:

$$\frac{\varphi^{n+1}-\varphi^{n}}{\Delta t} = f\left(\varphi^{n+1}\right) + N(z)\cdot\left(\varphi_0 - \varphi^{n+1}\right) \quad (16)$$

Here φ is an arbitrary model variable with $f(\varphi^{n+1})$ is all processes action on the variables predicted, n indicates the number of time step, $N(z)$ is the nudging weight, and φ_0 is the observed value, or an interpolated value between observations, of the model variable (Wang and Warner, 1988). For nudging we used data for temperature, specific humidity and wind speed measured at 4 different heights (8.4–74 m).

3.2 Emissions of volatile organic compounds

The emissions of organic vapours from the canopy were calculated with an approach based on MEGAN (Model of Emissions of Gases and Aerosols from Nature, Guenther et al., 2006). This model was coupled with SOSA to provide on-line estimates of landscape averaged emission rates of monoterpenes and other biogenic VOC's. The model estimates for a specific location are a function of leaf area, plant species composition and representative species-specific emission factors. Hourly variations in estimated emissions are driven by changes in calculated leaf temperature and incident direct and diffuse solar radiation on sun and shade leaves at different canopy levels.

3.3 Chemistry

All chemical reaction equations for the model runs were selected from the Master Chemical Mechanism (http://mcm.leeds.ac.uk/MCM/). The model runs included 2140 reactions with a total of 761 chemical species representing the complete reaction paths for isoprene, 2-methyl-3-buten-2-ol, β-pinene, α-pinene, methanol, acetone, acetaldehyde, formaldehyde, methane and all relevant inorganic reactions. KPP – the Kinetic PreProcessor (Damian, 2002; Sandu and Sanders, 2006) is used to translate the

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reaction equations into Fortran 90 code that performs the time integration of the kinetic system. Of the several numerical solvers for systems of differential equations available in KPP, we used the LSODE solver (Radhakrishnan and Hindmarsh, 1993; Sandu et al., 1997). The KPP-produced Fortran code is then called from main SOSA code. Some minimal changes to the KPP-produced code were performed to facilitate linking with main SOSA code.

The chemistry and meteorology are combined in a typical split-operator approach. Meteorology, including atmospheric mixing of the chemical species, is simulated with a 10 s time step and after 6 meteorology steps chemistry, separately for each atmosphere layer, is simulated for 60 s. Sensitivity studies with time steps from 5 to 300 s for the chemistry step showed that the selected time step of one minute is the longest interval with negligible effects on daily profiles of high and low reactivity compounds.

As the time integration of the system of 2140 differential equations for the kinetics was the most time consuming part of the model, the chemistry module was implemented with Message Passing Interface (MPI) library, so the chemistry steps for different atmospheric layers can be run on several cores on a computer or a computer cluster. In principle, all of the 75 atmospheric layers could be distributed to different cores, but in practice we noticed that after ca. 32 cores there was not much further speedup. The reason is, in some layers the time for solving the chemistry used by the solver (LSODE) is longer compared to others. Some cores manage to integrate 2 or 3 "fast" layer in the time some other core spends integrating one "slow" layer.

In total, simulating one month, using 75 atmospheric layers and the 2140 chemical reaction equations involving 761 chemicals, running SOSA on 32 cores on a cluster computer, typically took one hour.

3.4 Set up

The inputs required for the model calculations are of different types. The first group includes the characteristics of the area over which the calculations are performed. These include the vertical characteristics of the vegetation (structure, photosynthetic

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characteristics etc.). The second group consists of meteorological parameters (radiation conditions and vertical profiles of wind speed, temperature, moisture and passive scalars). The model is adjusted to use information regarding wind speed, temperature and moisture obtained from synoptic levels as initial boundary conditions for the upper border. For passive tracers, for example carbon dioxide, a typical value of its concentration above the mixed layer for given time of year is taken as the upper boundary condition. The last group of input values are the inorganic gases and the condensational sink. These parameters are measured at the SMEAR II and included every half hour with a linear integration in between.

The aerosol condensational sink (CS) determines how rapidly molecules will condense onto preexisting aerosols (Boy et al., 2003) and is calculated from

$$\mathrm{CS}=4\pi D\int_0^\infty r\beta_\mathrm{M}(r)n(r)dr=4\pi D\sum_i \beta_\mathrm{M} r_i N_i \quad (17)$$

Here r is the radius, N is the number concentration of the particles in the size class i and D is the diffusion coefficient of the condensing species. The transitional correction factor β_M is typically calculated using the expression by Hidy and Brock (1971). In this work we used the values for sulphuric and nitric acid to calculate the condensation sink values for both vapours with the assumption that nitric acid condenses onto particles at lower humidity (<60%) a thousand times slower but increases with increasing humidity to a value of ten percent compared to sulphuric acid at RH = 100%.

4 Results and discussion

4.1 Average canopy characteristics

The description of the turbulent structure of the atmospheric boundary layer using a one-dimensional model is necessarily a simplification of the real three-dimensional world. The approach works best for an open flat terrain or flat terrain with a uniform

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forest cover. Variations in topography or in forest composition will result in a more complex relationship between surface and canopy characteristics and quantities measured in a point over or inside the canopy. Our approach is to model the vertical leaf area density distribution using beta distribution, and then search for the shape of the distribution that produces the best match to observed turbulence statistics in and above the canopy in near-neutral conditions (Rannik et al., 2003).

Beta distribution has been used to represent profiles of leaf area density (e.g., Meyers and Paw U, 1986; Markkanen et al., 2003) or source distribution (e.g., Van den Hurk and McNaughton, 1995) inside the canopy in the form of

$$f\left(z/h;\alpha,\beta\right)=\frac{\left(z/h\right)^{\alpha-1}\left(1-z/h\right)^{\beta-1}}{\int_0^1\left(z/h\right)^{\alpha-1}\left(1-z/h\right)^{\beta-1}d\left(z/h\right)} \quad (18)$$

Two parameters, α and β, determine the shape of the distribution. When $\alpha>\beta$, the maximum value occurs where $z/h>0.5$, and therefore represents foliage (or a source) concentrated in the upper part of the canopy. For any set of non-negative α and β, $\int_0^{z/h} f\left(z/h\right)d\left(z/h\right)$ equals unity, thus the real leaf area density is the product of (18) and LAI/h. For given h, LAI and c_d, SOSA profiles matched the measured flow statistics best with values $\alpha=3$ and $\beta=3$.

The LAD profile used in our simulation (Fig. 1a) deviates slightly from that of Rannik et al. (2003). However, it should be remembered that the modelled profiles combine (or present the average of) canopy properties within the dynamical footprint of the tower, incorporating the effects of inhomogeneous vegetation and variations in topography. The agreement between measured and simulated mean flow statistics is more important here than the exact description of canopy architecture. The modelled flow and momentum flux match well with observations and are independent of C_μ (Fig. 1b and c). Although TKE and consequently velocity standard deviation vary with the choice of C_μ (Fig. 1d and e), the eddy diffusivity remains unaffected by its value (Fig. 1f), resulting in insensitivity of scalar fluxes and concentration distributions to the choice of C_μ in our model simulations.

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We should note that with a one-dimensional model where any advection both in horizontal and in vertical directions is ignored, the fitted vertical shape of LAD is representative only in an aerodynamic sense. This allows us to reconstruct successfully the flow dynamics at the tower location under neutrally stratified conditions. Reconstructions of vertical profiles of temperature and humidity that affect the flow under non-neutral conditions for such a complex area as Hyytiälä are mainly dependent on the nudging procedure.

4.2 Verification and test of approach

We demonstrate the performance of the model for two selected months in 2006. Figures 2 and 3 present temperature, friction velocity, sensible and latent heat fluxes, as measured and modelled for March and August 2006. The selected months can be considered as qualitatively and quantitatively representative of the whole year for these parameters. The simulated temperature profiles agree well with the measurements during daytime, and show an underestimation of the temperature decrease only on certain days during late evening and in the night. This behaviour of the model is visible especially on days with strong temperature gradients.

The modelled friction velocities follow the measurements in a satisfying manner. Taking uncertainties from the instrumentation and errors in the calculations of the friction velocity into account, SOSA predicts this parameter at nearly all times within the measurement accuracy. The comparisons for sensible and latent heat fluxes between the model and the measurements reflect on many days especially in summer an overestimation of downward fluxes (negative values) during nighttimes and on some days a similar but opposite behaviour (positive values) during daytimes.

Figures 2 and 3 demonstrate that the model is able to reproduce measured characteristics with a very good agreement most of the time. Some differences that are more pronounced between modelled and measured variables in particular days can be explained by the inability of the model to react to very fast changes of surrounding conditions such as passage of atmospheric fronts. Overestimation of negative fluxes

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in night conditions is mostly connected with the definition of R (long-wave radiation fluxes) in the model (see Eq. 3). Downward longwave flux depends on the emissivity of the atmosphere, which in turn is dependent on cloud cover fraction. Without records of cloudiness during the night the model, especially under calm wind conditions, is unable to reproduce heat fluxes properly. Also, the present version of the transport model does not include any influence of precipitation on atmospheric characteristics. Nevertheless, the model seems to be suitable for our purposes.

4.3 Model application

The newly developed model SOSA can be used for many possible applications of chemical-meteorological studies on different timescales. Problems like the missing OH-reactivity (e.g. Sinha et al., 2008), long-term relationships between certain gas-phase compounds and the formation processes of atmospheric particles, or tests of different emission models in comparison with measurements are all applications that will be addressed in future publications. In this paper we will focus on one type of organic species, monoterpenes, and will examine their vertical distribution and possible connections with the formation of very small particles below 6 nm.

Figure 4 presents the vertical profiles for the sum of the monoterpenes (α-pinene, β-pinene, D-limonene, D3-canrene, sabene and camphene), as averages for nighttime (01:00–04:00 a.m.) and daytime (10:00 a.m.–02:00 p.m.) between the 1 and 26 June 2007. Comparing the mean simulated concentrations between 4 and 22 m with the mean value measured by proton transfer reaction-mass spectrometry (PTR-MS, Rinne et al., 2005) at 4, 14 and 22 m resulted in marginal overestimations from the model by 14 and 8% for night- and daytimes, respectively (in the night, PTR-MS: 1.19×10^{10}, SOSA: 1.36×10^{10}; in the day, PTR-MS: 6.45×10^{9}, SOSA: 6.97×10^{9}; all values are in molecules cm^{-3}). During the day the profile of monoterpenes decreases with height inside the mixed layer (approximately 1 km thick on average in June) by about 60–70%. Above the ABL up to the height of 2.5 km a much stronger decrease to concentrations below 10^{6} molecules cm^{-3} is predicted. At nighttimes the modeled and

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measured concentrations are about doubled as compared to the daytime values near the surface, with a strong decline to one tenth at 300 m. In the nighttime residual layer the upward transport of molecules from the strong turbulent mixing in the daytime are partly dissolved through chemical, physical and meteorological mechanisms leading to a 60–80% reduction.

During the intensive field campaign of the OSOA (Origin and formation of Secondary Organic Aerosols) EU-project in Hyytiälä in August 2001 a total of 80 VOC samples were collected onto cartridges from tethered balloon platforms (Boy et al., 2004). The dispersion and reactions of the terpenes in this study resulted in a vertical gradient of -2.15×10^9 molecules cm^{-3} for the whole atmospheric boundary layer. Since not all of the necessary input data for SOSA are available for Hyytiälä for the years previous to 2003, we calculated an averaged vertical gradient of monoterpenes for 1 to 31 August 2007 and received a similar value with -2.75×10^9 molecules cm^{-3} for the ABL. Both comparisons with measurements – near the surface and the gradient inside the mixed layer – showed that SOSA simulations of the atmospheric concentrations of organic vapours (in this case the sum of monoterpenes) and their vertical profiles are in good agreement with measurements.

Observed vertical profiles throughout the troposphere of most parameters, like e.g. the size distribution of particles or organic vapour concentrations are rare because of the difficulty and expenses involved in the measurements. However, during the second intensive field campaign of the EU-project QUEST (Quantification of Aerosol Nucleation in the European Boundary layer, March–April 2003) the GTK Twin Otter Geophysics Research Aircraft (O'Dowd et al., 2007) and the microlight aircraft D-MIFU (Junkermann, 2005) were used for airborne studies. During the descent of the flight on the 26 March between 11:25–11:31 a.m., an inversion was located about 1100–1200 m. The surface mixed layer, below 600 m, exhibited large concentrations of nucleation mode particles while no nucleation mode particles were seen in the layer between 650–1100 m (O'Dowd et al., 2009).

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In Fig. 5 vertical profiles for the sum of monoterpenes, hydroxyl radicals and sulphuric acid predicted by SOSA for the 26 March 2003 at 11:25 are shown. Both compounds, H_2SO_4 and OH, present a similar trend with nearly constant values of approximately 4.5×10^7 molecules cm^{-3} and 7.5×10^5 molecules cm^{-3}, respectively, up to about 300 m. Above and up to 2150 m both compounds decrease by ca. 50% and start to increase afterwards. The sum of the monoterpenes showed a completely different profile, which is expected from a reactive substance emitted by the canopy. The concentration is well mixed inside the ABL – due to clear sky condition on this day and the resulting strong turbulence – with about 1.2×10^9 molecules cm^{-3} and decreases continuously to values below 100 molecules cm^{-3} above 2 km. Comparing the measured particle concentrations in the size range of 3–6 nm (high numbers of several thousand inside the ABL and numbers under the detection limit above, O'Dowd et al., 2009, Fig. 9) with the simulated profiles of sulphuric acid and monoterpenes, a higher similarity with the organic vapour vertical distributions could be identified. This could be strong evidence that the amount of newly formed particles detected at sizes above 3 nm are more related to the concentrations of organic molecules than to the concentrations of sulphuric acid inside and above the ABL.

Figure 6 shows the vertical distribution from the surface up to 1500 m for temperature, air moisture, eddy diffusivities for momentum and a scalar for the 26 March 2003, the same day as selected for Fig. 5. The temperature profile during the first 8 h shows an inversion in the residual layer between 200 and 400 m with about 3.5 K warmer air, as compared to the stable surface layer. Around 06:30 a.m. the mixed layer height starts to increase and in this context the inversion disappears and a strong temperature increase through solar heating leads to a maximum temperature inside the canopy at about one hour after local noon. The evolution of the ABL (white lines in Fig. 6a–d) reached its maximum at 01:00 p.m. at around 830 m and exactly at 11:25 the mixed layer height is predicted to be 604 m which is in excellent agreement with the measurements of the vertical particle profile by O'Dowd et al. (2009) as discussed above. The simulated development of the ABL is also visible in a strong upward transport of water

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molecules between 10:00–12:00 a.m. (see Fig. 6b). Both modelled quantities, temperature and air moisture reflect the inertia of the atmosphere after the breakdown of the ABL at around 03:00 p.m. For the temperature profile, a cooling at the surface after sunset and a slow but continuous upward heat flux above 300 m is predicted, whereas air moisture starts to decline slowly throughout the whole model domain for a short time after the ABL breakdown. The simulated evolution of the boundary layer gives the eddy diffusivity parameters for momentum or a scalar. At 310 m and 01:00 p.m. the maximum values are 44 and 140 $m^2 s^{-1}$ for momentum and a scalar, respectively.

5 Conclusions

During development of SOSA we have faced many problems related to requirements for scientific and technical aspects of modelling. Driven by scientific questions requiring a proper description of the transport processes within the ABL there were several technical problems that needed to be solved, namely what data is required and how it should be used in the model. The main problem is the decoupling between the conditions formed in the upper ABL or above the ABL (upper boundary conditions in our model), and local conditions formed near the surface. This manuscript presents a detailed description of the new SOSA model and demonstrates our modelling approach to reproduce a complete picture of turbulent flow inside and above a forest from discrete time measurements.

We clearly demonstrated that SOSA can reconstruct measured parameters very well and is able to reproduce meteorological parameters like the eddy diffusivity for momentum and for scalars, which can not be observed directly. The combination of a reliable meteorological parameterization with an emission model (MEGAN) for volatile organic compounds and a detailed chemical module has many possible applications. In particular, because of its parallelized implementation allowing for relatively fast run times, the new SOSA model allows for long term simulations of the biosphere-atmosphere interface with the best detailed chemistry available.

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The first application of SOSA presented in this manuscript focused on the vertical profiles of the sum of monoterpenes. These compounds are thought to have an important role in the formation and growth of particles over the boreal forest (Boy et al., 2003). Comparison with measured concentrations at the ground and in the ABL showed a good agreement with our model predictions. In response to the strong different opinions in the aerosol community about what molecules are responsible for the formation of new particles in the atmosphere (Boy et al., 2008) – organics or sulphuric acid – we compared vertical profiles for both compounds with measured particle concentrations between 3–6 nm. Although these results could not give any information about the processes involved, it is notable that a similar trend was observed between the sum of monoterpenes and the nucleation mode particles but no trend was found for sulphuric acid.

Ongoing projects with the new SOSA model include the calculation of OH-reactivity, long-term statistical analysis of different parameters in comparison with nucleation events and the test of different emission models in comparison with measurements. In addition the implementation of the University of Helsinki Multicomponent Aerosol code (UHMA, Korhonen et al., 2004) will give the possibility to test and improve different parameterizations for aerosol dynamical processes on longer time periods.

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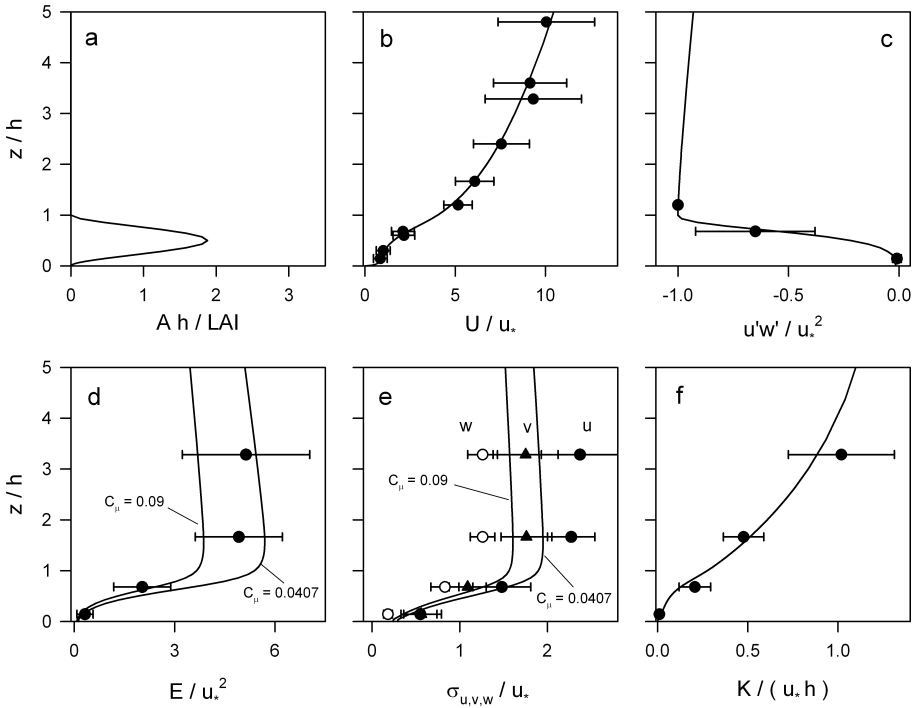

Fig. 1. Normalized vertical profile of foliage density **(a)** assumed in the model and normalized experimental (symbols) and modelled (lines) turbulence statistic within and above Pine Scots forest at Hyytiälä under near-neutral conditions: **(b)** the mean wind speed, **(c)** momentum flux, **(d)** turbulent kinetic energy, **(e)** standard deviation of wind speed components, and **(f)** eddy diffusivity. Error bars present ±1 standard deviation.

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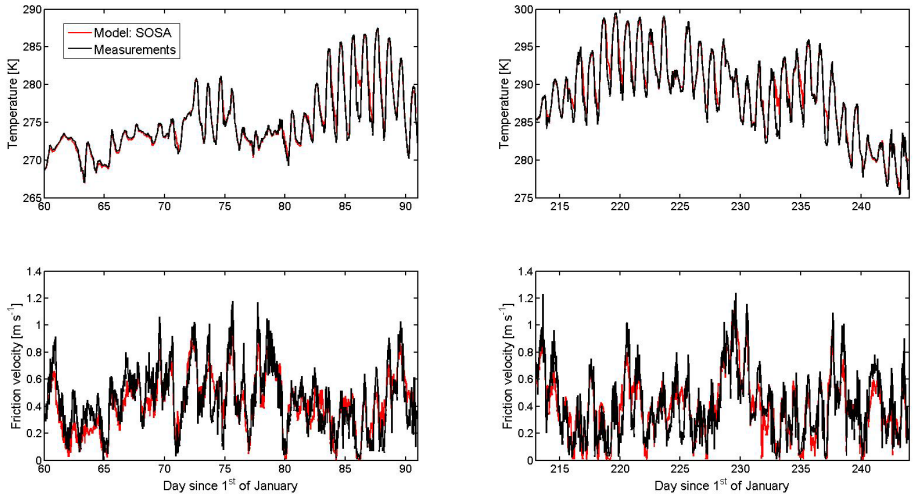

Fig. 2. Modelled and measured temperature and friction velocity for March and August 2007 for SMEAR II, Hyytiälä at 23 m above ground.

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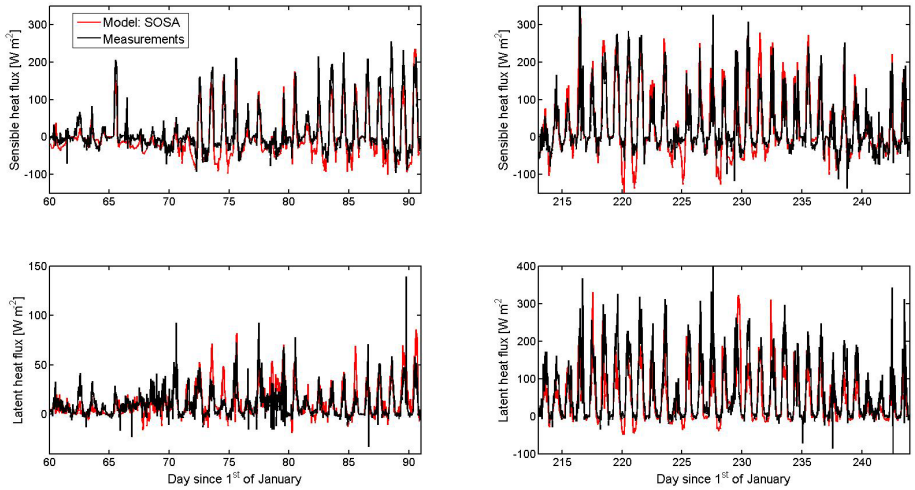

Fig. 3. Modelled and measured sensible and latent heat flux for March and August 2007 for SMEAR II, Hyytiälä at 23 m above ground.

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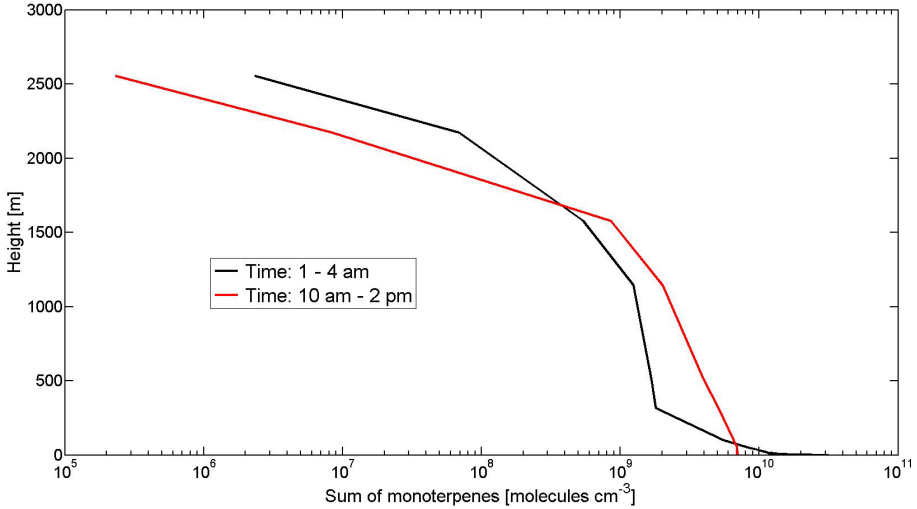

Fig. 4. Averaged vertical profiles of the sum of monoterpenes for the period from 1 to 26 June 2007 at two different time intervals.

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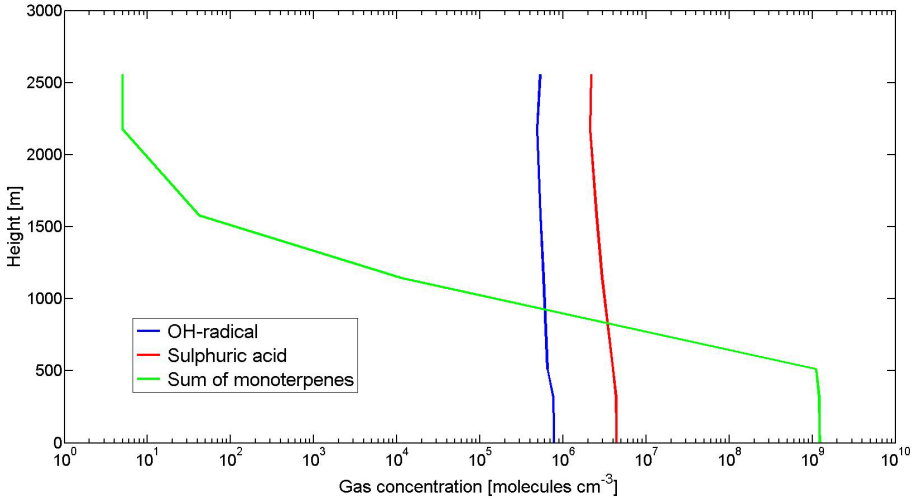

Fig. 5. Vertical profiles of OH-radical, sulphuric acid and sum of monoterpenes concentrations for 26 March 2003 at 11:25 a.m.

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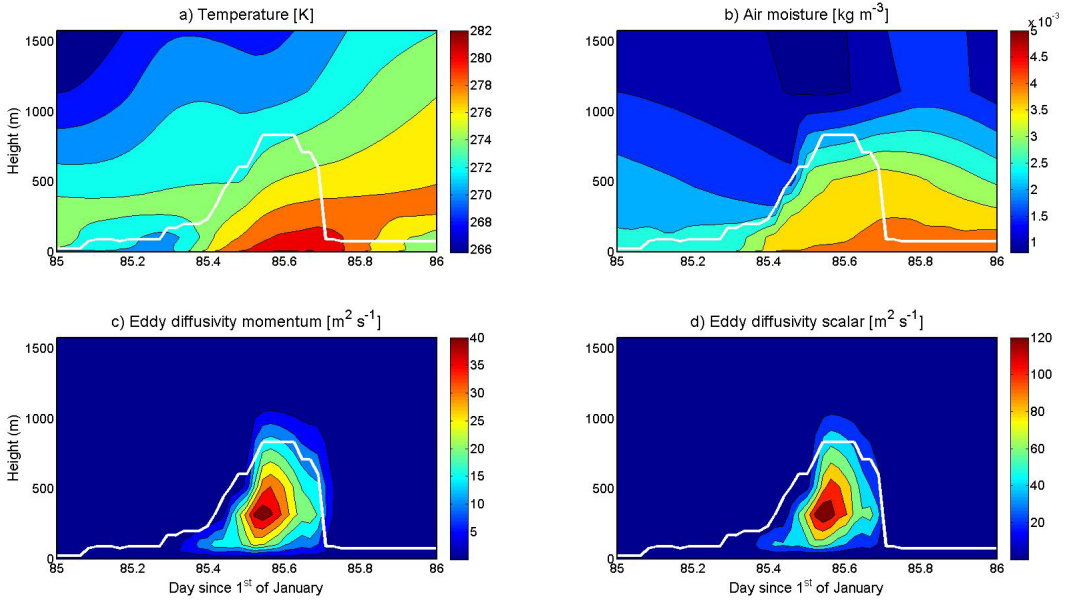

Fig. 6. Modelled vertical profiles of temperature, air moisture and eddy diffusivity for momentum and scalar for the 26 March 2003. The simulated evolution of the atmospheric boundary layer height is included by white line.

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