

Abstract

We studied the atmospheric boundary layer (ABL) dynamics and the impact on atmospheric chemistry during the HUMPPA-COPEC-2010 campaign. We used vertical profiles of potential temperature and specific moisture, obtained from 132 radio soundings, to determine the main boundary layer characteristics during the campaign. We propose a classification according to several main ABL prototypes. Further, we performed a case study of a single day characterized as a convective boundary layer to analyse the influence of the dynamics on the chemical evolution of the ABL, using a systematic analysis that can easily be extended to other periods during HUMPPA-COPEC-2010. We used a mixed layer model, initialized and constrained by observations. In particular, we investigated the role of large scale atmospheric dynamics (subsidence and advection) on the ABL development and the evolution of chemical species concentrations. We find that, if the large scale forcings are taken into account, the ABL dynamics are represented satisfactorily. Subsequently, we studied the impact of mixing with a residual layer aloft during the morning transition on atmospheric chemistry. The time evolution of NO_x and O_3 concentrations, including morning peaks, can be explained and accurately simulated by incorporating the transition of the ABL dynamics from night to day. We demonstrate the importance of the ABL height evolution for the representation of atmospheric chemistry. Our findings underscore the need to couple the dynamics and chemistry at different spatial scales (from turbulence to mesoscale) in chemistry-transport models and in the interpretation of observational data.

1 Introduction

The boreal forest, located roughly between 50° and 65° N, is an ecosystem type that covers 8% of the global land surface and 27% of the forested area, covering about $15 \times 10^6 \text{ km}^2$ (Williams et al., 2011). Therefore, its impact on the global atmospheric chemistry and physics is significant. Since it contains over 10% of the total carbon

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present in the combined ecosystems on Earth, perturbations in the climate of the Northern Hemisphere could lead to changes in the carbon cycle (including emissions of volatile organic compounds, VOCs) and aerosol formation and consequently alter the atmospheric composition (Sellers et al., 1997). Considering the extent of the boreal forest, perturbations in this ecosystem may alter the dynamics and chemistry at different temporal and spatial scales, up to the entire globe. Therefore, it is relevant and timely to study the processes in the boreal forest at a range of scales in order to understand how they interact and influence the atmospheric dynamics and chemistry.

During the HUMPPA-COPEC-2010 campaign (Williams et al., 2011), which took place at the Finnish SMEAR II station from 12 July to 12 August 2010, special emphasis was placed on obtaining a complete data set of surface and atmospheric measurements to comprehensively characterize the atmospheric physics and chemistry. Guided and constrained by this data, we focus on the influence of large scale forcings and transitions in the morning from nocturnal to daytime conditions on the boreal atmospheric boundary layer (ABL) dynamics and the associated atmospheric chemistry. Our research extends on the analyses of previous campaigns, like the Boreal Ecosystem-Atmosphere Study (BOREAS) (Sellers et al., 1997). BOREAS was conducted in the Canadian forest, aimed at improving the understanding of interactions between the boreal forest biome and the lower atmosphere (Sellers et al., 1997). Observations included dynamical, ecological and biogeochemical variables. The latter included observations of the trace gases CO₂, CH₄ and non-methane hydrocarbons. Even though the boundary layer dynamics were analysed (Barr and Betts, 1997; Davis et al., 1997), their evolution was not represented using models to identify and quantify acting processes. In addition, their impact on the atmospheric chemistry was not considered.

At the SMEAR II station in Hyytiälä (61°51' N, 24°17' E, 181 m a.s.l.) continuous observations are performed (Hari and Kulmala, 2005). The station is designed to study the transport of aerosols (Kulmala et al., 2001), gases, energy and momentum between the soil, the air within the canopy and the ABL. Due to the focus of SMEAR II, all obser-

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5 vations are performed in the soil, the canopy and the lower part of the ABL. Previous observational campaigns at this site focused on aerosol studies (Nilsson et al., 2001).

During the HUMPPA-COPEC-2010 campaign the standard instruments of the SMEAR II site were complemented with additional equipment. An overview of the gas and aerosol measurement instruments is given in Table 1 of Williams et al. (2011). To be able to characterize the ABL evolution, additional observations were made to obtain vertical profiles of the meteorological variables, using a Cessna aircraft and radiosondes. The campaign turned out to be of particular interest due to the anomalously high temperatures that might be representative of future boreal climates. Since the atmospheric temperature affects the surface latent and sensible heat fluxes (van Heerwaarden et al., 2010), future climates may be characterized by different ABL dynamics.

Concentrations of chemical species in the atmosphere are governed by surface exchange, chemical processes and dynamics in the ABL. Therefore, it is imperative to correctly represent the ABL dynamics when interpreting or predicting atmospheric chemistry (Davis et al., 1994; Ganzeveld et al., 2008; Vilà-Guerau de Arellano et al., 2011). A primary goal of our research is to determine how the growth of the ABL influences atmospheric chemistry. The ABL height development influences the chemical evolution of the boundary layer in two ways. First, the actual height of the ABL can be considered as a mixing volume in which reactive compounds are released. During the day, the height of this layer increases non-linearly from values as low as 100 m in the morning to values that can exceed 2 km in the afternoon. Second, the growth of the boundary layer determines the entrainment of air from the free troposphere into the boundary layer. This entrained air generally has different thermodynamic and chemical properties than air in the ABL.

25 In this paper we classify different boundary layer prototypes that correspond to the most representative vertical potential temperature profiles. This classification of the structure of the ABL enables the identification of important processes for atmospheric chemistry during HUMPPA-COPEC-2010, like the intensity of mixing, turbulent time

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scales, the atmospheric dilution capacity and the entrainment of free tropospheric air. An overview of these processes is given in Table 1 of Ouwersloot et al. (2011).

We further examine the boundary layer dynamics for a single day with particularly intensive observations, using a mixed layer model (MXL). Emphasis is placed on how atmospheric chemistry is affected by the ABL height evolution. The day is selected because important processes took place that can be easily overlooked when analysing the impact of the boundary layer dynamics on chemistry. These processes, associated with temporal transitions and large-scale forcings, include subsidence, the advection of air masses and, in the morning, the connection with a residual boundary layer of the previous day. We complete the study by evaluating the effectiveness of the applied observational strategy during HUMPPA-COPEC-2010 in characterizing the ABL dynamics.

The next section covers the methodology of this study. Subsequently, the representation of the boundary layer height during one representative day in the HUMPPA-COPEC-2010 campaign is presented. This is followed by an illustration of its importance for accurately modelling atmospheric chemistry. Finally, the strategy for obtaining meteorological data during field campaigns is discussed.

2 Methods

In this study, meteorological and atmospheric chemistry observations are combined with numerical experiments. Some observations serve as initial and boundary conditions, while others are compared to the numerical model results. The upper atmospheric conditions are determined by radiosondes and the atmospheric conditions near the Earth's surface are obtained at the SMEAR II measuring station. The used chemical observational techniques are described in Table 1 of Williams et al. (2011). We focus on the O₃ observations with the ultraviolet absorption/fluorescence (UV) method and the NO and NO₂ observations with the chemiluminescence (CLD) method, which

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were all performed by the Max Planck Institute for Chemistry at a height of 24 m above the ground.

Two complementary numerical techniques are used: the mixed layer model is applied to characterize the boundary layer dynamics and their possible effect on atmospheric chemistry, while vertical profiles generated in the numerical experiments with a Large-Eddy Simulation (LES) model act as virtual radio soundings for the discussion of the observed radiosonde profiles. More specific information is given in the following sections.

2.1 Radiosondes

During the campaign, 175 GRAW DFM-06 radiosondes have been launched from a clear area at approximately 300 m distance from the main observational site. In principle, five radiosondes were launched per day at the local daylight saving times (LT, UTC+3) 3:00, 9:00, 12:00, 15:00 and 21:00. The local daylight saving time is ahead of the local solar time by 1 h 29 min. During intensive observation periods (IOP), the frequency of radiosonde launches was increased to intervals of maximum two hours. These events took place from 16 July 21:00 to 17 July 18:00, from 21 July 21:00 to 22 July 18:00, from 28 July 21:00 to 29 July 18:00 and from 5 August 18:00 to 6 August 18:00. The observations of 6 August will be analysed in more detail.

Data transmitted by the sonde each second was received by an antenna placed at a nearby building. Next to a GPS device, each sonde included a temperature and a humidity sensor. The GPS coordinates were used to determine the altitude and geographical location of the sonde, as well as the wind velocities. The determined position is accurate within 10 m and the uncertainty of the wind velocity measurements is less than 0.1 ms^{-1} . The temperature sensor measured with an accuracy of 0.2°C and the humidity sensor measured the relative humidity with an accuracy of 1 %. The software of the ground station calculates the pressure profile by combining these observations with the surface pressure. From the combination of altitude, pressure, temperature and

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relative humidity data, the profiles of the potential temperature, θ , and specific humidity, q , are determined. This study focuses on the lowest 3000 m of these profiles.

2.2 SMEAR II data

Data near the surface was gathered at the boreal forest observational station SMEAR II (Hari and Kulmala, 2005). The standard observations include meteorological data, aerosols and the concentrations of several chemical species (CO_2 , H_2O , CO , O_3 , SO_2 , NO and NO_2). In addition, the turbulent fluxes of temperature, moisture, momentum, aerosols, CO_2 , O_3 and several volatile organic compounds are measured. For this study, the heat fluxes, surface pressure, relative humidity and temperature observations are used.

The sensible heat flux, H , and the latent heat flux, LE , are determined using 30-min averages over 10 Hz Eddy Covariance measurements at a height of 23 m (Mammarella et al., 2009). The observations were made above the canopy top, which was located at approximately 15 m height. As such they can be considered as appropriate surface forcings for the development of the ABL and can be directly related to the ABL dynamics observed from the radiosonde profiles.

The observational data of surface pressure and the relative humidity and temperature at 50 m height originate from the SMEAR smartSearch database (Junninen et al., 2009). These data are converted to potential temperature and specific humidity in a similar way as the radiosonde data.

2.3 Mixed layer model (MXL)

To represent and subsequently interpret the observational data, a mixed layer model is used that is coupled to a chemical module (Tennekes, 1973; Vilà-Guerau de Arellano et al., 2009). This model is based on the zeroth-order mixed layer assumption that during the day turbulent mixing is vigorous enough to result in a well-mixed ABL. Thus, the model is similar to a chemical box model with an evolving upper boundary. Mixed layer

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theory assumes that conserved scalar variables are uniformly distributed in the vertical direction and that their fluxes are therefore characterized by linear profiles. A thermal inversion layer caps the ABL and separates it from the free troposphere. Both the thermal inversion layer and the surface layer (lowest 10% of the boundary layer, Stull, 1988) are considered to be thin compared to the total boundary layer. This approach has first been used by Lilly (1968), Tennekes (1973), Carson (1973) and Betts (1973). The vertical profiles of the evaluated quantities show a discontinuity at the height of the inversion, above which they linearly change with height in the free troposphere according to their free tropospheric gradients. This simplified representation of the ABL, which nevertheless incorporates the processes under study, allows for numerical experiments in which atmospheric chemistry and basic ABL dynamics are simultaneously solved at very low computational costs. A model that is similar to the one used here, including chemistry, is described by Vinuesa and Vilà-Guerau de Arellano (2005). Further development includes expanding the chemical module and taking the influence of specific humidity on the entrainment rate into account (Vilà-Guerau de Arellano et al., 2009). The chemical module is flexible and enables the use of different chemical schemes. In previous work it has mostly been used to represent the O_3 - NO_x -VOC- HO_x system that is typical for the Amazonian rain forest. The currently applied chemical scheme, which is based on the scheme used by van Stratum et al. (2012), is presented in Table 1. The model acts as a support for the observational data, enabling to study the evolution of the main properties of the boundary layer dynamics.

More information about the governing equations is given in Appendix A.

2.4 LES model (DALES)

To evaluate the use of radio soundings to obtain vertical profiles of temperature and moisture, numerical experiments have been performed with the modified 3.2 version of the Dutch Atmospheric Large-Eddy Simulation (DALES) model (Heus et al., 2010). This model explicitly resolves dynamical (and chemical) processes in the boundary layer at grid resolutions finer than 100 m. Therefore, the resulting data can be compared to

local observations. In addition, DALES enables us to study how atmospheric flows that are induced by heterogeneous surface forcings influence the distribution of both thermodynamic and chemical variables.

This specific LES model has first been used by Nieuwstadt and Brost (1986) and further developed and improved since (e.g., Cuijpers and Duynkerke, 1993; Dosio et al., 2003). Processes are explicitly resolved on scales larger than a set filter width and parametrized on the smaller scales at which the eddies contain less energy. The resolved equations are the filtered Navier-Stokes equations upon which the Boussinesq approximation is applied (Heus et al., 2010). Typically, the filter width is chosen such that over 90 % of the turbulent energy is contained in the resolved scales. The subfilter-scale parametrization is based on the one-and-a-half-order closure assumption (Dear-
dorff, 1973). Periodic boundary conditions are applied in the horizontal directions.

The version of DALES used for this study is modified to enable studies of ABL flows characterized by heterogeneous boundary conditions at the surface (Ouwensloot et al., 2011) and to generate local instantaneous vertical profiles of the chemical species mixing ratios and the dynamical variables. This allows for a comparison between radiosonde profiles and the profiles that are predicted by numerical experiments

3 Results

3.1 Boundary layer prototypes

Our interpretation of the radiosonde observations indicates that during HUMPPA-COPEC-2010 no clear trend due to synoptic influences is present and that the characteristics of the ABL significantly differ each day. Therefore, the boundary layer dynamics and their influence on atmospheric chemistry have to be analysed on a day-to-day basis. To support this analysis, it is convenient to have a first classification of the ABL dynamics during HUMPPA-COPEC-2010 and associate the radiosonde measurements to boundary layer prototypes. By performing this classification, the observations can

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thus be linked to the representations in combined chemistry-meteorology models. The prototypes are characterized by specific vertical profiles and the associated dynamical processes (e.g., Stull, 1988). During the campaign, three different prototypes were observed that correspond to the most representative vertical potential temperature profiles. Out of the 175 radiosondes 43 could not be classified by these prototypes, either due to instrumental errors or different (transitional) characteristics in the observed vertical profiles. We select three profiles from the radiosonde observations to describe their main characteristics. The profiles are presented in Fig. 1.

Figure 1a shows the potential temperature profile observed at 16:00 LT on 17 July. It is characterized by a relatively constant potential temperature with height, topped by a layer of a few hundred meters in which the vertical gradient of the potential temperature gradually increases. The location of the maximum potential temperature gradient marks the height of the boundary layer (Sullivan et al., 1998), in this case at approximately 2150 m. From the top of the ABL, the potential temperature linearly increases with height, following the free tropospheric gradient. This increasing potential temperature profile indicates a region that is characterized by a stably stratified flow. The potential temperature is approximately equal in the entire boundary layer due to convective mixing. The jump in potential temperature at the top of the boundary layer, i.e. the thermal inversion, acts as a cap and limits the exchange of air between the ABL and the free troposphere. This measured profile is associated with the convective boundary layer that is usually formed by active convective turbulence during the day. As mentioned in Sect. 2.3, a commonly used approximation, the zeroth-order jump assumption (Garratt, 1992), assumes the inversion layer as infinitesimally small, resulting in a jump in the potential temperature at the top of the boundary layer. The resulting theoretical profile is plotted with dashed lines. This very common boundary layer prototype, especially in spring and summer, is evaluated further using the MXL model. ABL profiles of this kind are most clearly discernible during high pressure synoptic systems.

The profile of 18:00 LT on 13 July, as presented in Fig. 1b, is similar to the previously discussed one, but contains an extra layer. Until a height of 1050 m the potential

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temperature is again constant. However, in this case this mixed layer is topped with a conditionally unstable layer. This type of ABL is normally observed with active shallow cumulus (fair weather) clouds (Stull, 1988). The conditionally unstable layer is located between the mixed layer and the inversion layer and is characterized by an increase with height of the potential temperature that is stronger than the dry and weaker than the moist adiabatic lapse rate. The dry adiabatic lapse rate is defined as the increase of potential temperature that an unsaturated rising air parcel would experience under adiabatic conditions, while the moist adiabatic lapse rate describes this increase for a saturated rising air parcel. In short, the conditionally unstable layer acts as a turbulence suppressing stable layer for unsaturated air parcels and as a turbulence generating unstable layer for saturated air parcels. In the first case (clear air), this layer can be considered as part of the thermal inversion layer, while in the latter case (clouds) air parcels that enter the layer rise to its top. When this profile was observed, the humidity was too low to result in condensation (below 70 % relative humidity) and clouds. If under such conditions condensation would have occurred for local air parcels in the conditionally unstable layer, the resulting clouds would become active and grow. Above the conditionally unstable layer, the thermal inversion is located at a height of 2500 m. Above the inversion, the potential temperature rises according to the free tropospheric profile. The corresponding boundary layer prototype, shown by the black dashed line, is a mixed layer topped with a conditionally unstable layer.

The third radiosonde profile is depicted in Fig. 1c and is based on data obtained at 3:00 LT on 7 August. It shows a potential temperature that increases with height in a layer that reaches up to a few hundred meters above the surface. Aloft the potential temperature is relatively constant with height. On top, a thermal inversion at 1700 m and a free tropospheric profile are present. This data is characteristic for nocturnal stable boundary layers that are formed after an afternoon transition (e.g., Kaimal et al., 1976; Nieuwstadt and Brost, 1986). During the evening transition and at night, the air near the surface cools, driven by the emission of longwave radiation. This leads to stratification and a stable boundary layer. A residual mixed layer of the previous day can

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be present above the stable boundary layer, as is the case in this example. However, this is not the case for all stable boundary layers and residual layers can disappear as time progresses (Kaimal et al., 1976). After sunrise, the Earth's surface warms and convective turbulence mixes the air. The stable boundary layer characteristics are then dissipated and a new convectively mixed boundary layer is formed. If a residual layer is still present, the new mixed layer develops until the potential temperature is equal in both layers. When this happens, buoyant thermals that originate at the surface enter the residual layer without passing through a stable atmospheric layer, a process called overshooting. It results in an almost instantaneous mixing of the new boundary layer with the air masses in the residual layer. From that moment on, mixed layer theory can be applied to study the evolution of the combined ABL. This process of connecting a shallow boundary layer with a residual layer is depicted in Fig. 2. Its impact will be analysed in more detail in Sect. 3.4, putting special emphasis on the evolution of the concentrations of the chemical species NO_2 and O_3 .

An overview of the occurrence of all three boundary layer prototypes during HUMPPA-COPEC-2010 is given in Table 2. Note that the radiosonde profiles in Fig. 1 are selected based on their unambiguous structure. In general, due to the instantaneous measurements, deviations of the radiosonde profile compared to the domain averaged profile occur, caused by different atmospheric processes. One of these, which is often overlooked, is the influence of land surface heterogeneity on the distribution of temperature, moisture and chemical species. To show that these deviations are realistic, Fig. 3 presents local instantaneous potential temperature profiles as generated by DALES. This simulation is based on the period between 12:00 and 15:00 LT of the MXL case that is treated in more detail in Sect. 3.2. The resolution of the domain is 50 m in the horizontal direction and 20 m in the vertical direction. The numerical grid spans $256 \times 256 \times 175$ cells in, respectively the x , y and z directions, resulting in a domain of $12.8 \text{ km} \times 12.8 \text{ km} \times 3.5 \text{ km}$. Constant surface heat and moisture fluxes are imposed for 3 h after which the local instantaneous profiles are generated. The kinematic surface heat flux and moisture flux are 0.24 K ms^{-1} and $0.087 \text{ g kg}^{-1} \text{ ms}^{-1}$, corresponding to

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a sensible and latent heat flux of 295 W m^{-2} and 262 W m^{-2} , respectively. In case of heterogeneous surface forcings, the domain is split into two patches in the x -direction, the *dry* and the *wet* patch. The kinematic surface heat flux is increased (decreased) by 0.04 K m s^{-1} and the kinematic surface moisture flux is decreased (increased) by $0.017 \text{ g kg}^{-1} \text{ m s}^{-1}$ for the *dry* (*wet*) patch. It appears from Fig. 3a that for the homogeneous surface forcings, even though the domain averaged profile is smooth (inset), the individual profiles slightly differ from each other and show more random behaviour due to the turbulent character of the ABL, i.e. local warm and cold parcels of air. As demonstrated by the blue dashed line, this can result in observed boundary layer heights that differ from the domain average. Figure 3b shows that the differences between local instantaneous profiles are enhanced for heterogeneous surface forcings, especially over the patch with the higher sensible heat flux. This is due to the generated mesoscale circulations (Ouwersloot et al., 2011) in the ABL. These local differences should be kept in mind during the analysis of the radiosonde profiles.

3.2 Diurnal evolution of the ABL

To describe the boundary layer dynamics in greater depth, a specific day is selected, characterized by a boundary layer of the mixed layer type. The chosen day, 6 August 2010, was scheduled as an IOP, so multiple radio soundings were performed at relatively short time intervals (every 2 h) during the day. The profiles of θ and q show clear mixed layer prototype behaviour (Fig. 1a) for these soundings, which enables the analysis of the data using the MXL model. The day is of interest for the interpretation of chemical observations, since most instruments were functioning and the wind came from the south-west, which was its predominant direction (Table 2 of Williams et al., 2011). Therefore, chemical data is available for this day under conditions that are typical for the campaign. Closely related to our research questions, additional dynamical processes are of importance, though easily overlooked in the interpretation of the impact of ABL dynamics on atmospheric chemistry, as will be discussed below. These

processes include advection of air masses, subsidence and the coupling of a dynamically evolving boundary layer with a residual layer aloft during the morning transition. The advection and subsidence are both related to forcings on the meso- and synoptic scale. Note that under typical conditions for which subsidence occurs (high pressure and temperature), the emissions of primary biogenic compounds are usually larger (e.g., Guenther et al., 2006; Yassaa et al., 2012).

Figure 4 shows the time evolution of the boundary layer dynamics as observed by the radiosondes. The vertical profiles of potential temperature and specific humidity are displayed in Fig. 4a and 4b, respectively. For clarity, only four profiles (at 6:00, 10:00, 14:00 and 18:00 LT) are presented out of the eight available ones (at 6:00, 8:00, 10:00, 12:00, 14:00, 16:00, 18:00 and 21:00 LT). We omit the radiosonde profiles near the surface (until 100–300 m) due to the poor reception of the sonde data by the antenna of the ground station if the sonde is at lower heights. At higher altitude, the signal is less perturbed and data is received regularly. In Fig. 4c, the evolution of the boundary layer height is presented. Boundary layer heights are denoted by stars, while the top of the residual layer (in the morning) is denoted by circles.

To initialize the MXL model, the vertical profiles of the potential temperature and specific humidity for an initial point in time are applied. For this study, the residual layer in the observations at 8:00 LT is chosen as initial profile. Since the residual layer is much deeper than the underlying shallow mixed layer (975 m and 150 m, respectively) and both layers connect and mix within the first hours of the day (between 8 and 10 LT), considering the residual layer as a start of a new mixed layer seems a valid approximation.

The radiosonde observations are also used to derive the effects of large scale subsidence (downward moving air) and advection. These effects can not be observed directly, but by using the radiosonde observations as a reference for the predicted evolution of the ABL dynamics, one can obtain a first order estimation. To determine the subsidence velocity, the observed growth of the boundary layer height is compared to the modelled evolution velocity due to entrainment. The subsidence velocity is calculated

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as the difference between these two velocities, as expressed by Eq. (A4). To derive the contribution of the horizontal advection of air masses we take a different approach. The horizontal advection of air, with different dynamical and/or chemical properties, results in an increase or decrease in the mixed layer averaged scalars. In our model this rate of change is considered to be constant in time. Therefore, a difference between the observed and modelled scalar values that increases linearly in time is considered to be caused by advection. For the case under study we find that the advection of cooler air is of importance for temperature (with a cooling rate of 0.108 K h^{-1} resulting in a maximum difference of 1.5 K), whereas there is no significant contribution of advection to humidity. This approach cannot be directly applied to the advection of chemical species, since the free tropospheric concentrations are unknown. In addition, differences can be caused by unaccounted chemical pathways.

The prescribed sensible and latent heat fluxes in the MXL model are determined by fitting sinusoids through the observed fluxes, as shown in Fig. 5a. The complete set of initial conditions and forcings is presented in Table 3. In Fig. 5b–d, the simulated evolutions of the main dynamical ABL characteristics are shown together with the observations. This shows that the model reproduces the radiosonde observations for temperature, moisture and boundary layer heights well. Until approximately 17:00 LT, entrainment most strongly influences the ABL height development, resulting in a deepening ABL. When the boundary layer reaches its maximum height of 1740 m, the subsidence velocity for that height is equal in magnitude as the entrainment velocity. By that time the entrainment process is still very active, as the driving heat fluxes are still stronger than 50% of their maximum values. Since the surface sensible and latent heat fluxes continuously decrease after 14:00 LT, the entrainment fluxes weaken as well. Consequently, after 17:00 LT the subsidence becomes stronger than the rate of the boundary layer growth by entrainment and the ABL height decreases with time. This can be seen in Fig. 5b. The potential temperature, shown in Fig. 5c, increases with time due to surface fluxes and the related entrainment of relatively warm air from the free troposphere. After sunset, the surface buoyancy flux fades and, as a result, the

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convective boundary layer type (Fig. 1a) is not longer applicable. A stable boundary layer appears with a residual layer aloft (Fig. 1c). During the whole day, the advection of relatively cool air results in a decrease of the potential temperature. After 18:00 LT this effect is stronger than the heating effect due to surface fluxes and entrainment, explaining the temperature decline. As shown by the time evolution in Fig. 5d, the specific humidity remains approximately constant, since the effects of the surface moisture flux and the entrainment of relatively dry air cancel each other ($\frac{\partial(q)}{\partial t} \approx 0$).

To determine the importance of advection and subsidence, four different cases have been simulated. The three cases other than the previously derived standard MXL case differ by having the subsidence and/or advection disabled. We omit the effect of subsidence by setting $\text{Div}_U = 0 \text{ s}^{-1}$ and the horizontal advection of air with a different temperature by setting $\text{Adv}_\theta = 0 \text{ K s}^{-1}$ (Appendix A). First we discuss the impact of subsidence. From Fig. 6a we find that subsidence is significant for the boundary layer height development on 6 August 2010. In the cases without subsidence the boundary layer grows like a standard convectively mixed boundary layer until approximately 20 LT. Subsequently, the surface heat flux does not remain positive and the growth stops. Since this layer is no longer actively mixed, it remains as a residual layer on top of the stable nocturnal boundary layer. When subsidence occurs, the boundary layer growth slows down, resulting in lower ABL heights. As mentioned earlier, after 18:00 LT the boundary layer height decreases as the subsidence is stronger than the ABL height increase due to entrainment and the ABL becomes shallower. The rate at which the boundary layer height decreases reaches its maximum when entrainment ceases. The subsidence does not influence the incoming heat fluxes, but does result in shallower boundary layers. Therefore, subsidence promotes a warmer ABL. This is discernible in Fig. 6b.

Figure 6a shows that the advection of relatively cold air from other regions does not affect the boundary layer height evolution. The reason is that the advection is assumed to be equal in the free troposphere and the boundary layer, hence the strength of the thermal inversion remains virtually unaltered. The difference in potential temperature due to advection increases linearly in time up to 1.5 K after the 14 h of simulated time,

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as depicted in Fig. 6b. During the day, the impacts of advection and subsidence on the potential temperature roughly cancel in the case under study. Therefore, if both effects are ignored, the resulting potential temperature evolution is similar to that when both effects are concurrently considered. Our results indicate that the effects of both subsidence and horizontal advection are important to take into account.

3.3 Importance of the ABL height representation for atmospheric chemistry

After determining the evolution of the ABL dynamics, we study how it influences the mixing ratios of reactive species. Here we focus on the importance of a correct representation of the boundary layer height. Note that we solely study the primary effect of the boundary layer height evolution on the mixing ratio of chemical species, i.e. the ratio between surface exchange and mixing height (first term on the r.h.s. of Eq. A1). To prevent biases due to a combination of dilution and entrainment (e.g., Vilà-Guerau de Arellano et al., 2011), the initial mixing ratios are set equal in the boundary layer and the free troposphere. Two atmospheric chemistry cases are evaluated to show the impacts of the ABL dynamics and surface emissions depending on the region under study. For the first case the initial conditions and surface forcings are based on boreal conditions, while a second case is formulated to contrast the boreal forest to the tropical Amazon rainforest conditions. The initial mixing ratios, in ppbv, and the emissions at the surface, in $\text{mg m}^{-2} \text{h}^{-1}$, are set to 0, except for the values listed in Table 4.

We designed three different numerical experiments: a “*realistic*” case, a *constant* case and a case that considers the residual layer part of the free troposphere (by *ignoring the residual layer*). The three corresponding boundary layer height evolutions are shown in Fig. 7a. The first case corresponds with that described in the previous section, though expanded with the chemistry calculations. Thus the boundary layer height evolution is equal to the one presented above, which represents the observations. The second case represents a boundary layer with a constant height, mimicking the performance of chemical box models. For the third case, the initial profile is taken from the radiosonde at 8:00 LT, but the difference between the residual layer and the

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free troposphere aloft is ignored. Therefore, this layer is considered to be part of the free troposphere only and the prescribed initial mixed layer conditions and the free tropospheric gradients both change. The effect of subsidence is not considered for this situation. The altered settings for this case are listed in Table 5.

In Fig. 7b the resulting evolutions of the hydroxyl radical (OH) mixing ratio are presented for the boreal conditions. Since the emissions of isoprene (C_5H_8) are relatively low, and its background concentration is negligible, the depletion of OH is governed by other chemical species (especially CO). As we prescribe uniform initial profiles in the ABL and the free troposphere and neglect surface exchanges for these other chemical species, the resulting time evolutions of OH are very similar. As presented in Fig. 7c, it becomes different for our case with Amazonian conditions. In this case, the initial concentration and the emission of isoprene are higher and the depletion of OH due to isoprene is significant. During the first six hours of simulated time, if the emitted isoprene is distributed over a smaller mixing volume, the isoprene concentrations become higher and, consequently, the depletion of OH is enhanced. The *constant* boundary layer height case, which has the larger mixing volume, therefore starts with the highest OH concentration. The ABL height and OH mixing ratio are lowest for the case that ignores the residual layer. Due to the additionally produced secondary reactants (e.g. through RO_2) this effect remains even after the boundary layers reach the same height for two cases. Because of this non-linear effect, the OH mixing ratio remains highest in the case with the constant boundary layer height and lowest in the case where the influence of the residual layer is ignored. Our findings show that for an adequate model representation of atmospheric chemistry in the boundary layer, it is important to represent the boundary layer height evolution throughout the day. This holds especially for cases in which emitted chemical compounds significantly affect the chemical conditions.

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3.4 Representation of the morning transition

As discussed in Sect. 3.1, in the early morning a residual mixed layer of the previous day is frequently observed above the stable boundary layer and the shallow boundary layer that follows when convection starts. Due to the initially weak surface heat fluxes, the potential temperature in the shallow boundary layer increases until it is similar to the residual mixed layer (in our case around 9:30 LT). Then both layers merge and become one mixed boundary layer, as illustrated in Fig. 2. In the MXL model this merging of two atmospheric layers into one is instantaneous. However, in reality it can take some time due to inhomogeneous ABL conditions and the time it takes for thermals to rise from the surface to the top of the residual layer. In this section we will show that this process can explain specific patterns in the chemistry observations during the morning transition. Note that these numerical experiments do not aim for a perfect representation of the boreal atmospheric chemistry, though demonstrate that this merger of two atmospheric layers is an important process that may explain certain features in the observations of chemical species in the morning.

Three numerical experiments were designed. The first, the *standard* case, corresponds to that previously defined in Sect. 3.2, except for the chemical conditions. This case assumes the residual layer to be part of the mixed layer. The second experiment, the *lower layer*, is based on the initial shallow boundary layer. This corresponds to situation (1) in Fig. 2. The initial values for potential temperature, specific humidity and ABL height are again obtained from the radiosonde observation of 8:00 LT. This numerical experiment is run for 1.5 h. After that the buoyant thermals that originate from the shallow boundary layer enter the residual layer and the two layers mix almost instantaneously, resulting in situation (2) in Fig. 2. The third numerical experiment represents the ensuing combined mixed layer. This case is referred to as *connected layers* and is labelled situation (3) in Fig. 2. The dynamical settings that differ per numerical experiment are listed in Table 6 for all three cases. Dynamical settings not mentioned explicitly are equal to the case prescribed in Sect. 3.2 and Table 3. The prescribed

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emissions and initial mixing ratios, presented in Table 7 if non-zero, are chosen such that the observations of the isoprene, NO, NO₂ and O₃ mixing ratios are reproduced.

The observed and modelled mixing ratios of NO₂ and O₃ are presented in Fig. 8a and 8b, respectively. The time evolution of NO (not shown) has similar features as that of NO₂. The observations show that in the early morning (until 9:30 LT) the NO₂ mixing ratio rises quickly. This is due to the emission of NO_x into the shallow boundary layer. Subsequently, the two atmospheric layers merge. Since the air in the residual layer has lower mixing ratios of NO and NO₂, the mixing ratios in the combined mixed layer are lower than the mixing ratios in the previous shallow boundary layer. Because in reality the conditions of the ABL are not completely horizontally homogeneous, this mixing does not occur simultaneously everywhere, resulting in a transition that takes approximately one hour. After the mixing of the two layers, the NO_x contributions of emission and entrainment are still positive, but are distributed over a larger mixing volume. Therefore, their impact becomes weaker and, due to chemical destruction by OH, the NO₂ mixing ratio slightly decreases with time. The morning peak in NO₂ can not be reproduced by the MXL model if only one numerical experiment is performed. However, by combining two numerical experiments, one for the shallow boundary layer and one for the final combined mixed layer, this peak can be reproduced and explained. It is worthwhile to note that the longer the separation between shallow boundary layer and residual layer remains present, the stronger this morning peak is. Additionally, due to storage in the canopy under a stable boundary layer, NO_x could accumulate near the surface during the night. When turbulence sets in, the canopy and ABL air masses interact, which could cause pulses in the surface exchange in the early morning (Ganzeveld et al., 2002).

The O₃ mixing ratio in Fig. 8b does not show a sudden decrease or increase. However, it is apparent that the time evolution changes after the morning transition from a shallow to a merged boundary layer. The increase in O₃ over time is not driven by surface exchange but mainly by entrainment. In the shallow boundary layer the O₃ mixing ratio is lower than in the residual layer. Therefore, entrainment causes the mixing

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ratio to rise in the growing shallow boundary layer. During the transition both layers are combined into one mixed layer. The rate of change of O_3 in this merged boundary layer is altered due to two reasons: the mixing ratio in the atmospheric layer above the original residual layer is different and the entrained air is mixed over a larger mixing volume after the combination. In effect this results in a slower increase of the O_3 mixing ratio. Note that the assumption of one well-mixed boundary layer from the start enables us to get a reasonable first approximation of the mixing ratio evolution. However, the representation in the morning is off and the differences in the rate of change of O_3 before and after 9:30 LT can not be explained. By using the first order approximation to consider in succession the *lower layer* and *connected layers* cases, the time evolution of O_3 can be explained and reproduced.

Our findings show that by using a relatively basic modelling tool, the mixed layer model, and using a first order approximation for the combining of a shallow boundary layer with a residual layer aloft, important features in the morning observations of chemical species can be reproduced and explained. We therefore conclude that the interpretation of atmospheric chemistry observations using a numerical model requires that dynamical processes are accounted for, including the boundary layer height evolution and the connection to a residual layer. This supports previous budget studies using the MXL model (Vilà-Guerau de Arellano et al., 2011; van Stratum et al., 2012) and a single column model (e.g., Ganzeveld et al., 2008).

4 Reflection on the observational strategy during HUMPPA-COPEC-2010

In this section, we extend the previous analysis to formulate recommendations for the observational strategy during campaigns such as HUMPPA-COPEC-2010. The experiences during the HUMPPA-COPEC-2010 campaign enable us to improve the strategy for future campaigns, resulting in an even more complete and comprehensive set of atmospheric data.

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As presented in Fig. 3, local observations can result in variations of the measured scalars under the influence of the boundary layer dynamics. For the potential temperature, the largest variations occur in the surface layer and near the inversion layer. The variability of the scalars is even enhanced by heterogeneous surface forcings (Ouwersloot et al., 2011). Thus, we conclude that for representative observations of the boundary layer dynamics it would be recommendable to launch multiple radiosondes simultaneously at key moments (e.g., morning transition, noon and evening transition) to account for the influences of heterogeneous terrain and local, instantaneous observations. In addition, these multiple radiosondes would provide important information about the spatial variations in the dynamical variables, which could be used to evaluate results of numerical models, including large scale chemistry-transport models.

Furthermore, the lowest few hundred meters of the atmosphere are important to characterize in detail. Within a convective boundary layer a surface layer is present in which the profiles of potential temperature, specific humidity and concentrations of chemical species can have significant vertical gradients. The nocturnal boundary layer, characterized by stable stratification, has a similar height as well. However, the observations during HUMPPA-COPEC-2010 did not suffice to fully characterize this part of the ABL. The observations at the towers are only performed at six fixed locations below 67 m and the radiosonde profiles near the surface are occasionally omitted due to poor transmission of the sonde data. To enable the characterization of the nocturnal boundary layer and the surface layer during the day, it is recommended to operate kytoons (tethered balloons) (Stull, 1988) with the relevant instruments for at least these lower areas. Due to the tether, no data will be lost and the speed of the ascend/descend during the profiling can be controlled.

As discussed in Sect. 3.2, another possible improvement to the employed observational strategy is to obtain information about large scale forcings, i.e. the subsidence velocity and horizontal advection of air masses. The information about subsidence could be estimated by comparing the boundary layer heights for subsequent observed profiles. This could be achieved by launching radiosondes within very short time intervals

(maximum 1 h). After calculating the entrainment velocity from the observed inversion layer properties and surface heat fluxes (Eq. A5), it can be subtracted from the observed ABL growth rate to determine the subsidence velocity (Eq. A4). In combination with the horizontal wind direction and velocity, the advection can be calculated from the horizontal gradients of the different scalars (Eq. A1). The influence of horizontal advection for potential temperature and specific humidity can therefore be determined by simultaneously launching at least 3 radiosondes around the observational site to obtain the horizontal distribution of these scalars. This approach has previously been applied to tower observations (e.g., Aubinet et al., 2003).

As presented in Fig. 4c, the observations during HUMPPA-COPEC-2010 enable a characterization of the development of ABL dynamics. However, for most days observations were limited to 5 radiosonde launches. During the intensive observation periods additional measurements were performed at relatively short time intervals, but even then continuous measurements are not available. A continuous representation of the ABL height would be relevant input for chemical box models as discussed in Sect. 3.3 and demonstrated in Fig. 7. Continuous observations could be obtained using a ceilometer, sodar (Nilsson et al., 2001) or a lidar (Gibert et al., 2007). This would also enable that the morning transition can be studied in greater detail, and more specifically the mixing of a shallow boundary layer with a residual layer aloft. Another advantage of continuous observations would be that fluctuations in the measurements due to local observations can be filtered by time averaging the data, according to Taylor's frozen turbulence hypothesis (Stull, 1988).

5 Conclusions

By combining observations, both near the surface and in the free troposphere, with a mixed layer model, we studied the atmospheric boundary layer dynamics as observed over the Finnish boreal forest during the HUMPPA-COPEC-2010 campaign. Particular emphasis is placed on the role of atmospheric dynamics at different scales

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on the boreal atmospheric chemistry. We investigated the influence of large scale forcings (subsidence and advection) and the transition from nocturnal to daytime turbulent conditions on the development of the ABL.

The meteorological data has been classified by identifying boundary layer prototypes based on the vertical potential temperature profiles. During the campaign three different types were observed: the stable boundary layer, the convectively mixed boundary layer and the conditionally unstable layer above a mixed layer. Of these three types, the convective boundary layer was observed most often, 78 out of the 132 classified soundings. Illustrated by Large-Eddy Simulation model results, we discuss how instantaneous observed profiles can deviate from these prototypes.

By selecting a single day, characterized by a convective boundary layer, 6 August 2010, we studied in detail the key dynamic contributions that influence atmospheric chemistry. This analysis could be applied to other cases observed during HUMPPA-COPEC-2010. It is shown that by using a relatively basic numerical model, the mixed layer model (MXL), the evolution of the boundary layer dynamics can be reproduced and explained. A residual mixed layer in the early morning and the effects of two different large scale forcings, subsidence and horizontal advection, have been shown to be important. During this day, the horizontal advection of cold air results in a decrease of the temperature at the measurement site. Subsidence inhibited the boundary layer growth, causing a lower boundary layer height and consequently, due to a smaller mixing volume and unaltered sensible and latent heat fluxes, higher temperatures. By accounting for both subsidence and cold air advection, the modelled evolution for temperature was shown to remain approximately equal, though the resulting atmospheric boundary layer height evolution was significantly reduced.

It is demonstrated that the representation of atmospheric chemistry with a numerical model requires accurate knowledge of the ABL height evolution. Due to the dependency of the chemical reaction rates on the concentrations of chemical species, knowledge about the initial (early morning) chemical conditions, the surface emissions/deposition and the boundary layer height at one specific point in time does

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not suffice to accurately predict the concentrations of chemical species at that point in time.

The morning transition from a shallow boundary layer, merging with a residual mixed layer aloft into a combined mixed boundary layer, has been represented by combining two numerical experiments with the mixed layer model. The results show that by using this assumption, we are able to explain and represent an observed morning peak in the NO_x concentrations and the increase of O_3 concentrations with time.

By using the mixed layer model, several processes are identified that require attention during observational field campaigns if measured data is to be reproduced. We find that emphasis should be placed on continuous observations of the atmospheric boundary layer height, combined with model analyses. Using this combined information, the effects of mixing with the residual layer and the entrainment and dilution of chemical species can be evaluated. A continuous representation could be achieved by using radiosondes and a numerical model, although this method assumes a boundary layer that is characterized by the convective boundary layer prototype and requires knowledge of the large scale forcings. Therefore, we suggest the use of a ceilometer, sodar or lidar in future campaigns.

Appendix A

Mixed layer equations

The prognostic equations are solved by the mixed layer model for multiple conserved scalar variables, S . These variables are the potential temperature, θ , specific humidity, q , and the mixing ratio's of chemical species, c_{species} . θ and q influence the evolution of the ABL height. Since the ABL is well mixed during the day, the averages of the variables over the whole boundary layer, $\langle S \rangle$, can be used for the evaluation. The difference between the free tropospheric and boundary layer values is symbolized by ΔS . The system of prognostic equations that solves $\langle S \rangle$ and ΔS is expressed by

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(Vilà-Guerau de Arellano et al., 2009):

$$\frac{\partial \langle S \rangle}{\partial t} = \frac{\overline{w'S'_s} - \overline{w'S'_e}}{h} - \left(U \frac{\partial \langle S \rangle}{\partial x} + V \frac{\partial \langle S \rangle}{\partial y} \right) + R_S, \quad (\text{A1})$$

$$\frac{\partial \Delta S}{\partial t} = \gamma_S w_e - \frac{\partial \langle S \rangle}{\partial t}. \quad (\text{A2})$$

An illustration of the different contributions to the boundary layer height development and to the prognostic equation for the boundary layer averaged scalars is presented in Fig. 9. The different terms are explained in more detail below.

$\overline{w'S'_s}$ and $\overline{w'S'_e}$ are the surface and entrainment fluxes, respectively. The height of the mixed layer is h . U and V are the wind velocities in the x and y direction and the total term $-\left(U \frac{\partial \langle S \rangle}{\partial x} + V \frac{\partial \langle S \rangle}{\partial y} \right)$ is the advection of S , Adv_S . The advection is positive if it leads to an increase in S . R_S expresses additional sources or sinks for scalar S . In the case of a chemical species, R_S is equal to the chemical production minus the chemical destruction of that species, based on the applied chemical mechanism. The vertical gradient in the free troposphere is expressed by γ_S . The velocity at which free tropospheric air is entrained into the boundary layer is symbolized by w_e .

The surface fluxes are prescribed, though for the zeroth-order jump assumption the entrainment flux at the top of the mixed layer is expressed by

$$\overline{w'S'_e} = -w_e \Delta S. \quad (\text{A3})$$

Next to the scalar variables, the boundary layer height evolution is included by applying

$$\frac{\partial h}{\partial t} = w_e + w_s. \quad (\text{A4})$$

Here, w_s is the subsidence velocity, associated with large scale forcings. It is related to the boundary layer height by $w_s = -\text{Div}_U h$, where Div_U is the velocity divergence in

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s^{-1} . Note that this represents a linear profile with $w_s = 0$ at the surface, as presented in Fig. 9b. The entrainment velocity is diagnosed by solving

$$w_e = -\frac{\overline{w'\theta'_{ve}}}{\Delta\theta_v} \quad (\text{A5})$$

together with the closure assumption that $\overline{w'\theta'_{ve}} = -\beta\overline{w'\theta'_{vs}}$, where usually $\beta = 0.2$.

5 The virtual potential temperature, θ_v , that appears in this set of equations, is equal to

$$\theta_v = \theta (1 + 0.61q), \quad (\text{A6})$$

which results in

$$\overline{w'\theta'_v} = (1 + 0.61\overline{q})\overline{w'\theta'} + 0.61\overline{\theta}\overline{w'q'}, \quad (\text{A7})$$

$$\Delta\theta_v = \Delta\theta + 0.61(\langle q \rangle \Delta\theta + \langle \theta \rangle \Delta q + \Delta\theta \Delta q). \quad (\text{A8})$$

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Table 1. Chemical reaction scheme solved by the MXL model, based on van Stratum et al. (2012). T is the absolute temperature in K and χ is the solar zenith angle. The unit of first-order reaction rate coefficients is s^{-1} and that of second-order reaction rate coefficients is $\text{cm}^3 \text{molec}^{-1} \text{s}^{-1}$. Reaction (R15), (R24) and (R25) have more comprehensive expressions for their reaction rates^{1,2,3}. For these expressions, c_{air} and $c_{\text{H}_2\text{O}}$ are the concentrations in molec cm^{-3} for air and H_2O , respectively. Reactions (R1), (R5) and (R6) are photolysis reactions. MVK is the acronym of methyl vinyl ketone and REST stands for products and/or secondary fast reactions with reactants in the ambient air that are not specified.

Name	Chemical equation	Reaction rate constant
R1	$\text{O}_3 + h\nu \rightarrow \text{O}^1\text{D} + \text{O}_2$	$3.83 \times 10^{-6} \cdot e^{-\frac{6575}{T}}$
R2	$\text{O}^1\text{D} + \text{H}_2\text{O} \rightarrow 2 \text{OH}$	$1.63 \times 10^{-10} \cdot e^{-\frac{60}{T}}$
R3	$\text{O}^1\text{D} + \text{N}_2 \rightarrow \text{O}_3 + \text{REST}$	$2.15 \times 10^{-11} \cdot e^{-\frac{110}{T}}$
R4	$\text{O}^1\text{D} + \text{O}_2 \rightarrow \text{O}_3$	$3.30 \times 10^{-11} \cdot e^{-\frac{60}{T}}$
R5	$\text{NO}_2 + h\nu \rightarrow \text{NO} + \text{O}_3 + \text{REST}$	$1.67 \times 10^{-2} \cdot e^{-\frac{6575}{2T}}$
R6	$\text{CH}_2\text{O} + h\nu \rightarrow \text{HO}_2 + \text{REST}$	$1.47 \times 10^{-4} \cdot e^{-\frac{6575}{2T}}$
R7	$\text{OH} + \text{CO} \rightarrow \text{HO}_2 + \text{CO}_2 + \text{REST}$	2.40×10^{-13}
R8	$\text{OH} + \text{CH}_4 \rightarrow \text{CH}_3\text{O}_2 + \text{REST}$	$2.45 \times 10^{-12} \cdot e^{-\frac{1775}{T}}$
R9	$\text{OH} + \text{C}_2\text{H}_6 \rightarrow \text{RO}_2$	1.00×10^{-10}
R10	$\text{OH} + \text{MVK} \rightarrow \text{HO}_2 + \text{CH}_2\text{O} + \text{REST}$	2.40×10^{-11}
R11	$\text{HO}_2 + \text{NO} \rightarrow \text{OH} + \text{NO}_2$	$3.50 \times 10^{-12} \cdot e^{-\frac{500}{T}}$
R12	$\text{CH}_3\text{O}_2 + \text{NO} \rightarrow \text{HO}_2 + \text{NO}_2 + \text{CH}_2\text{O} + \text{REST}$	$2.80 \times 10^{-12} \cdot e^{-\frac{500}{T}}$
R13	$\text{RO}_2 + \text{NO} \rightarrow \text{HO}_2 + \text{NO}_2 + \text{CH}_2\text{O} + \text{MVK}$	1.00×10^{-11}
R14	$\text{OH} + \text{CH}_2\text{O} \rightarrow \text{HO}_2 + \text{REST}$	$5.50 \times 10^{-12} \cdot e^{-\frac{105}{T}}$
R15	$2 \text{HO}_2 \rightarrow \text{H}_2\text{O}_2 + \text{O}_2$	k^1
R16	$\text{CH}_3\text{O}_2 + \text{HO}_2 \rightarrow \text{REST}$	$4.10 \times 10^{-13} \cdot e^{-\frac{500}{T}}$
R17	$\text{RO}_2 + \text{HO}_2 \rightarrow \text{REST}$	1.50×10^{-11}
R18	$\text{OH} + \text{NO}_2 \rightarrow \text{HNO}_3$	$3.50 \times 10^{-12} \cdot e^{-\frac{100}{T}}$
R19	$\text{NO} + \text{O}_3 \rightarrow \text{NO}_2 + \text{O}_2$	$3.00 \times 10^{-12} \cdot e^{-\frac{1000}{T}}$
R20	$\text{OH} + \text{HO}_2 \rightarrow \text{H}_2\text{O} + \text{O}_2$	$4.80 \times 10^{-11} \cdot e^{-\frac{500}{T}}$
R21	$\text{OH} + \text{H}_2\text{O}_2 \rightarrow \text{H}_2\text{O} + \text{HO}_2$	$2.90 \times 10^{-12} \cdot e^{-\frac{100}{T}}$
R22	$\text{NO} + \text{NO}_3 \rightarrow 2 \text{NO}_2$	$1.80 \times 10^{-11} \cdot e^{-\frac{110}{T}}$
R23	$\text{NO}_2 + \text{O}_3 \rightarrow \text{NO}_3 + \text{O}_2$	$1.40 \times 10^{-13} \cdot e^{-\frac{2470}{T}}$
R24	$\text{NO}_2 + \text{NO}_3 \rightarrow \text{N}_2\text{O}_5$	k^2
R25	$\text{N}_2\text{O}_5 \rightarrow \text{NO}_2 + \text{NO}_3$	k^3
R26	$\text{N}_2\text{O}_5 + \text{H}_2\text{O} \rightarrow 2 \text{HNO}_3$	2.50×10^{-22}
R27	$\text{N}_2\text{O}_5 + 2 \text{H}_2\text{O} \rightarrow 2 \text{HNO}_3 + \text{H}_2\text{O}$	1.80×10^{-39}
R28	$\text{HO}_2 + \text{O}_3 \rightarrow \text{OH} + 2 \text{O}_2$	$2.03 \times 10^{-16} \cdot \left(\frac{T}{300}\right)^{4.57} \cdot e^{-\frac{600}{T}}$

$$^1 k = (k_1 + k_2) \cdot (1 + k_3), \quad k_1 = 2.2 \times 10^{-13} \cdot e^{-\frac{600}{T}}, \quad k_2 = 1.9 \times 10^{-33} \cdot e^{-\frac{980}{T}} \cdot c_{\text{air}}, \quad k_3 = 1 + 1.4 \times 10^{-21} \cdot e^{-\frac{2200}{T}} \cdot c_{\text{H}_2\text{O}},$$

$$^2 k = 0.35 \cdot (k_1 \cdot k_2) / (k_1 + k_2), \quad k_1 = 3.6 \times 10^{-30} \cdot \left(\frac{T}{300}\right)^{-4.1} \cdot c_{\text{air}}, \quad k_2 = 1.9 \times 10^{-12} \cdot \left(\frac{T}{300}\right)^{0.2},$$

$$^3 k = 0.35 \cdot (k_1 \cdot k_2) / (k_1 + k_2), \quad k_1 = 1.3 \times 10^{-3} \cdot \left(\frac{T}{300}\right)^{-3.5} \cdot e^{-\frac{11000}{T}} \cdot c_{\text{air}}, \quad k_2 = 9.7 \times 10^{14} \cdot \left(\frac{T}{300}\right)^{0.1} \cdot e^{-\frac{11080}{T}}.$$

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Table 2. Occurrence of atmospheric boundary layer prototypes during HUMPPA-COPEC-2010 according to the radiosonde observations. Listed are the local daylight saving times (UTC+3). Times printed in bold correspond to radiosonde profiles used in this study. The three distinguished prototypes are the mixed layer, ML, the mixed layer topped with a conditionally unstable layer, UC, and a stable boundary layer, SL.

Type	Time of observation					
ML	09:00, 12 Jul	15:00, 12 Jul	21:00, 12 Jul	09:00, 13 Jul	12:00, 13 Jul	
	12:00, 15 Jul	15:00, 15 Jul	09:00, 16 Jul	12:00, 16 Jul	15:00, 16 Jul	
	21:00, 16 Jul	22:00, 16 Jul	23:00, 16 Jul	00:00, 17 Jul	01:00, 17 Jul	
	02:00, 17 Jul	03:00, 17 Jul	04:00, 17 Jul	10:00, 17 Jul	12:00, 17 Jul	
	14:00, 17 Jul	16:00, 17 Jul	18:00, 17 Jul	21:00, 17 Jul	09:00, 18 Jul	
	21:00, 18 Jul	09:00, 19 Jul	21:00, 19 Jul	12:30, 20 Jul	15:15, 21 Jul	
	21:00, 21 Jul	00:00, 22 Jul	09:00, 22 Jul	10:00, 22 Jul	12:00, 22 Jul	
	14:00, 22 Jul	18:00, 22 Jul	21:00, 22 Jul	03:00, 23 Jul	09:00, 23 Jul	
	12:00, 23 Jul	15:00, 23 Jul	03:00, 24 Jul	16:00, 24 Jul	21:00, 24 Jul	
	03:00, 25 Jul	12:00, 26 Jul	21:00, 26 Jul	09:00, 27 Jul	12:00, 27 Jul	
	21:00, 27 Jul	12:00, 28 Jul	15:00, 28 Jul	12:00, 29 Jul	14:00, 29 Jul	
	16:00, 29 Jul	18:00, 29 Jul	21:00, 30 Jul	09:00, 01 Aug	15:00, 02 Aug	
	18:00, 02 Aug	12:00, 03 Aug	18:00, 03 Aug	12:00, 04 Aug	16:00, 04 Aug	
	00:00, 05 Aug	09:00, 05 Aug	12:00, 05 Aug	10:00, 06 Aug	12:00, 06 Aug	
	14:00, 06 Aug	16:00, 06 Aug	18:00, 06 Aug	21:00, 06 Aug	12:00, 07 Aug	
	15:00, 08 Aug	09:00, 10 Aug	18:00, 10 Aug			
	UC	15:00, 13 Jul	18:00, 13 Jul	12:00, 18 Jul	15:45, 18 Jul	15:00, 20 Jul
21:00, 20 Jul		13:30, 21 Jul	22:00, 21 Jul	23:00, 21 Jul	12:00, 24 Jul	
15:00, 27 Jul		09:00, 30 Jul	15:00, 30 Jul	15:00, 31 Jul	21:00, 31 Jul	
12:00, 01 Aug		18:00, 01 Aug	18:00, 04 Aug	22:00, 04 Aug	15:00, 05 Aug	
18:00, 05 Aug		20:00, 05 Aug	22:00, 05 Aug	15:00, 07 Aug	18:00, 08 Aug	
SL	03:00, 12 Jul	03:00, 13 Jul	21:00, 13 Jul	03:00, 14 Jul	09:00, 14 Jul	
	16:00, 14 Jul	03:00, 18 Jul	03:00, 19 Jul	04:00, 22 Jul	05:00, 22 Jul	
	06:00, 22 Jul	21:00, 25 Jul	03:00, 26 Jul	09:00, 26 Jul	03:00, 27 Jul	
	21:00, 28 Jul	22:00, 28 Jul	00:00, 29 Jul	02:00, 29 Jul	06:00, 29 Jul	
	08:00, 29 Jul	03:00, 01 Aug	03:00, 02 Aug	11:00, 04 Aug	14:00, 04 Aug	
	06:00, 06 Aug	08:00, 06 Aug	03:00, 07 Aug	21:00, 08 Aug		



Table 3. Overview of the prescribed initial and boundary conditions. Presented are the initial values (subscript 0) for h , θ and q , the initial differences between the free troposphere and boundary layer (additional prefix Δ) and the free tropospheric gradients (denoted by γ) for θ and q , the surface fluxes (subscript s), the large scale divergence, Div_U , and the advection of heat, Adv_θ , and moisture, Adv_q , for the MXL model. The time, t , is expressed in hours local daylight saving time. The MXL model is run for 14 h.

Property	Value
t_0 (h)	8
h_0 (m)	975
Div_U (s^{-1})	1.5×10^{-5}
$\langle \theta \rangle_0$ (K)	292.5
$\Delta \theta_0$ (K)	0.5
γ_θ (K m^{-1})	4.7×10^{-3}
Adv_θ (K s^{-1})	-3×10^{-5}
q_0 (g kg^{-1})	8
Δq_0 (g kg^{-1})	-0.5
γ_q ($\text{g kg}^{-1} \text{ m}^{-1}$)	-2.7×10^{-3}
Adv_q ($\text{g kg}^{-1} \text{ s}^{-1}$)	0
$\rho c_p \overline{w' \theta'_s}$ (W m^{-2})	$310 \left(\frac{1}{2} - \frac{1}{2} \cos \left(2\pi \frac{t-6.5}{14.5} \right) \right)$
$\rho L_v \overline{w' q'_s}$ (W m^{-2})	$275 \left(\frac{1}{2} - \frac{1}{2} \cos \left(2\pi \frac{t-5.75}{16.25} \right) \right)$

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Table 4. Overview of the prescribed initial mixing ratios (ppbv) and the emissions at the surface ($\text{mg m}^{-2} \text{h}^{-1}$) of the reactive species for Sect. 3.3 for both boreal and Amazonian chemical conditions. Not listed are the values for species without emissions and zero initial mixing ratio and the values for O_2 and N_2 . These latter two species have constant mixing ratios of 2×10^8 and 8×10^8 ppbv, respectively. For the emissions, the functions $f_1 = (\frac{1}{2} - \frac{1}{2} \cos(2\pi \frac{t-4}{18}))$ and $f_2 = \sin(2\pi \frac{t-4}{18})$ can be used with t in hours local daylight saving time. The conversion factors for the NO and C_5H_8 emissions from ppbvms^{-1} to $\text{mgm}^{-2}\text{h}^{-1}$ are 4.4 and 10, respectively.

	O_3	NO	NO_2	CH_4	CH_2O	MVK	C_5H_8	CO	H_2O_2
Boreal chemical conditions									
Initial mixing ratio	24.5	0.07	0.224	1759	0.171	0.0877	0.17	101.95	0.0266
Prescribed emission	0	$1.76 \times 10^{-2} f_1$	0	0	0	0	$0.1 f_2$	0	0
Amazonian chemical conditions									
Initial mixing ratio	10	0	1	1724	0	1.3	2	124	0
Prescribed emission	0	2.2×10^{-3}	0	0	0	0	6.5	0	0

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Table 5. Overview of the altered initial and boundary conditions for the case in which the residual layer is ignored compared to the standard case in Table 3. Listed are initial values (subscript 0), free tropospheric gradients, γ , and the large scale velocity divergence, Div_U .

Property	Value
h_0 (m)	100
Div_U (s^{-1})	0
$\langle \theta \rangle_0$ (K)	290
$\Delta \theta_0$ (K)	1
γ_θ (K m^{-1})	6.3×10^{-3}
q_0 (g kg^{-1})	9
Δq_0 (g kg^{-1})	0
γ_q ($\text{g kg}^{-1} \text{m}^{-1}$)	-2.2×10^{-3}

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Table 6. The prescribed initial values (subscript 0) and free tropospheric gradients, γ , for three different numerical experiments with the MXL model. The initial time, t_0 , is expressed in hours local daylight saving time and the runtime of the model is expressed by t_{run} in hours.

Property	Standard run	Lower layer	Connected layers
t_0 (h)	8	8	9.5
t_{run} (h)	14	1.5	12.5
h_0 (m)	975	150	1027
$\langle \theta \rangle_0$ (K)	292.5	290.0	292.73
$\Delta \theta_0$ (K)	0.5	2.0	0.73
γ_θ (K m ⁻¹)	4.7×10^{-3}	1.5×10^{-3}	4.7×10^{-3}
q_0 (g kg ⁻¹)	8	9	8.06
Δq_0 (g kg ⁻¹)	-0.5	0	-0.91
γ_q (g kg ⁻¹ m ⁻¹)	-2.7×10^{-3}	-2.2×10^{-3}	-2.7×10^{-3}

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Table 7. The prescribed initial mixing ratios (ppbv) and the emissions at the surface ($\text{mgm}^{-2}\text{h}^{-1}$) of the reactive species for the numerical experiments discussed in Sect. 3.4. The conversion factors for the NO and C_5H_8 emissions from ppbvms^{-1} to $\text{mgm}^{-2}\text{h}^{-1}$ are 4.4 and 10, respectively. The initial conditions are listed for the boundary layer, BL, and the atmospheric layer aloft, TOP. Maximum emissions are denoted by EM and emission patterns by PAT. COS corresponds to an emission of $\text{EM}(\frac{1}{2} - \frac{1}{2}\cos(2\pi\frac{t-4}{18}))$ and SIN to an emission of $\text{EM}\sin(2\pi\frac{t-4}{18})$. Not listed are the values for species without emissions and zero initial mixing ratio and those for CH_4 , O_2 and N_2 . These latter three species have constant mixing ratios of 1759 , 2×10^8 and 8×10^8 ppbv, respectively.

	O_3	NO	NO_2	CH_2O	MVK	C_5H_8	CO	H_2O_2
Standard run								
BL:	24.5	0.07	0.224	0.171	0.0877	0.170	101.95	0.0266
TOP:	52.0	0.10	0.500	0.000	0.0000	0.000	101.95	0.0000
EM:	0.0	1.76×10^{-2}	0.0	0.0	0.0	0.1	0.0	0.0
PAT:	–	COS	–	–	–	SIN	–	–
Lower layer								
BL:	24.5	0.07	0.224	0.171	0.0877	0.170	101.95	0.0266
TOP:	38.0	0.19	0.500	0.000	0.0000	0.000	101.95	0.0000
EM:	0.0	1.76×10^{-2}	0.0	0.0	0.0	0.1	0.0	0.0
PAT:	–	COS	–	–	–	SIN	–	–
Connected layers								
BL:	35.0	0.07	0.26	0.645	0.0765	0.176	101.95	0.0841
TOP:	47.0	0.10	0.42	0.000	0.0000	0.000	101.95	0.0000
EM:	0.0	1.76×10^{-2}	0.0	0.0	0.0	0.1	0.0	0.0
PAT:	–	COS	–	–	–	SIN	–	–

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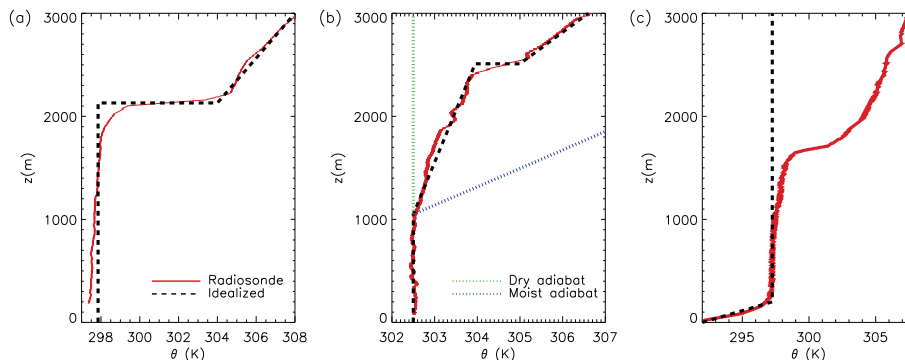


Fig. 1. Characteristic vertical potential temperature profiles for **(a)** a mixed layer, **(b)** a mixed layer topped with a conditionally unstable layer and **(c)** a stable boundary layer. In red an example from the radiosondes is presented, while in black an idealized profile is depicted. In **(b)** the profile of the potential temperature in the conditionally unstable layer is situated between the dry adiabat and the moist adiabat. In **(c)** a residual mixed layer of the previous day is present. The data is obtained at **(a)** 16:00 LT on 17 July, **(b)** 18:00 LT on 13 July and **(c)** 3:00 LT on 7 August.

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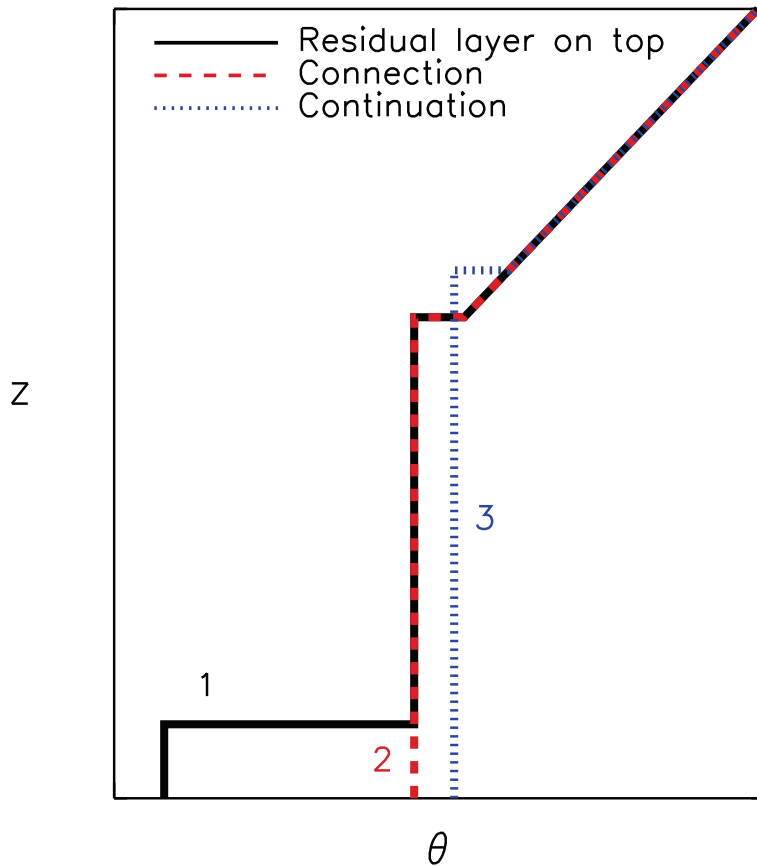


Fig. 2. Vertical potential temperature profiles during a transition from a mixed boundary layer topped with a residual boundary layer to a combined ABL. Initially **(1)** a shallow boundary layer develops under the influence of surface heat fluxes. When the potential temperatures in this layer and in the residual layer are equal, both layers connect **(2)**. After both layers are connected, the combined ABL further develops as usual **(3)**.

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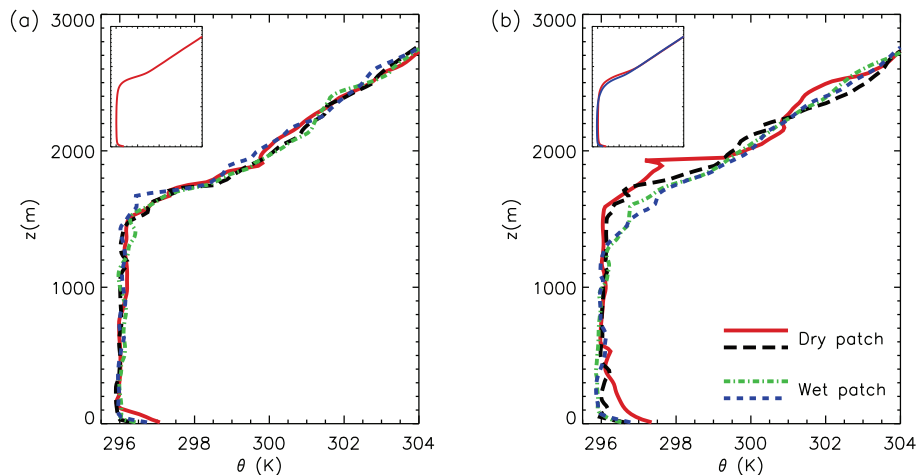


Fig. 3. Instantaneous and local potential temperature profiles as generated by DALES after 3 h of simulation. Panel (a) shows 4 profiles at different locations, while its inset shows the instantaneous horizontal average over the total domain. In panel (b), 2 instantaneous profiles are displayed for the dry patch and 2 profiles for the wet patch. Its inset presents the horizontal averages over the dry and wet patch separately.

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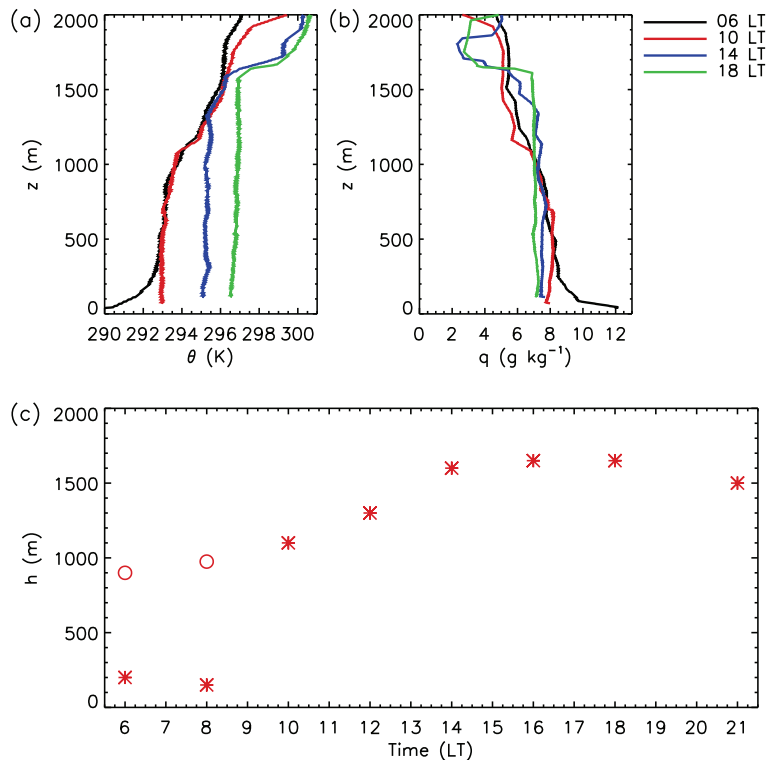


Fig. 4. Evolution of the vertical profiles of **(a)** the potential temperature and **(b)** specific humidity as measured by radiosondes on 6 August 2010 (6:00, 10:00, 14:00 and 18:00 LT). In **(c)** the evolution of the boundary layer height is presented, derived from all vertical profiles observed during that day. Stars indicate the height of the boundary layer, while circles represent the height of the residual layer in the morning. The shallow boundary layer connects with the residual layer between 8 and 10 LT.

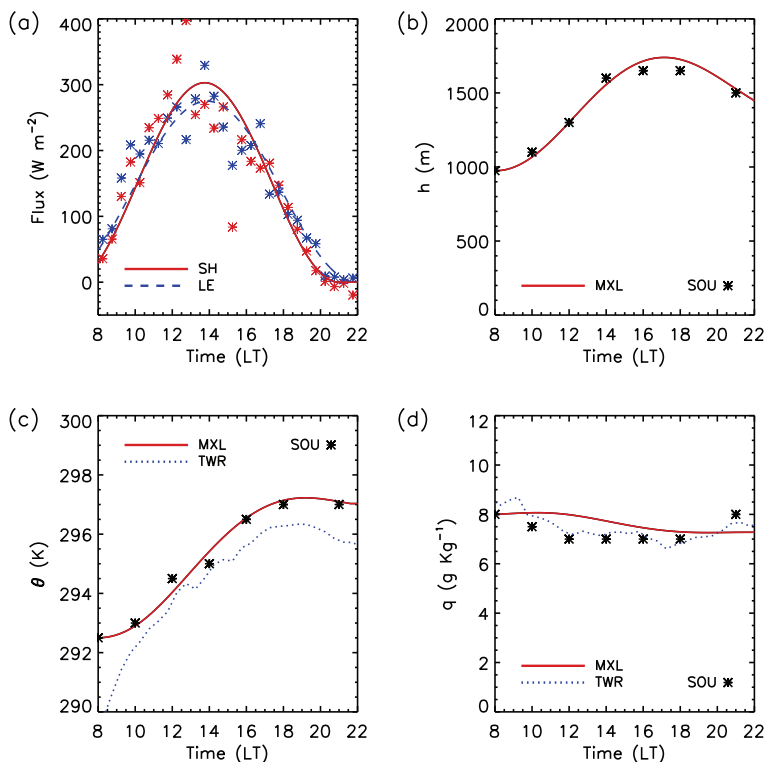


Fig. 5. Applied surface forcings **(a)** and simulated evolution of ABL characteristics **(b, c, d)** by the MXL model. The ABL characteristics include the boundary layer height **(b)**, potential temperature **(c)** and specific humidity **(d)**. In **(a)**, the lines represent the sensible heat flux, SH, and latent heat flux, LE, surface forcings applied to the model and the stars represent the corresponding observations from the tower. In **(b, c, d)**, solid red lines depict model results and the stars symbolize radiosonde observations. Tower observations (TWR) are expressed by dotted blue lines.

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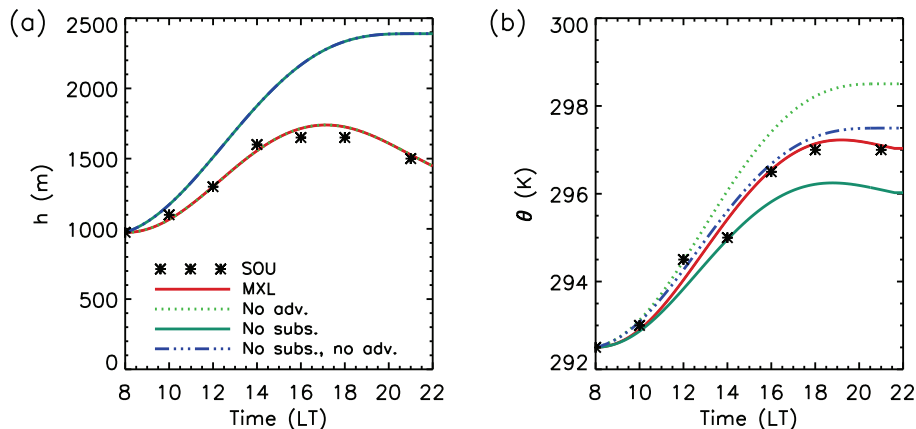


Fig. 6. Different evolutions of **(a)** the ABL height and **(b)** the potential temperature predicted by the MXL model. Subsidence and advection are separately enabled and disabled for the different numerical experiments, resulting in four cases. The results are compared to radiosonde observations, SOU. The most accurate case, with advection and subsidence enabled, is labelled MXL. Note that the advection does not significantly affect the boundary layer height evolution. Therefore, the curves are superposed in **(a)** for the cases with and without prescribed advection.

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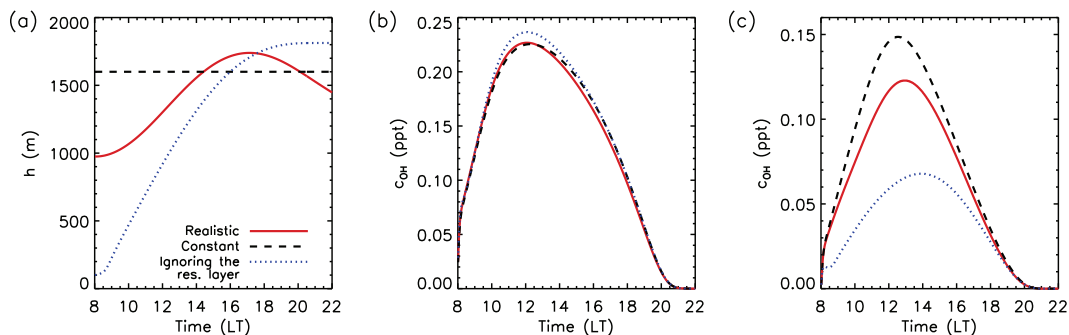


Fig. 7. Impact of **(a)** the ABL height development on **(b, c)** the mixing ratio of OH. Panels **(b)** and **(c)** present the results for initial chemical conditions and surface exchanges typical for boreal (as in HUMPPA-COPEC-2010) and Amazonian (Ouwersloot et al., 2011) conditions, respectively. Three cases are presented: (1) the boundary layer height as determined by the MXL model for 6 August 2010, (2) a constant boundary layer height and (3) a boundary layer height as determined by the MXL model if the residual layer would be considered as part of the free troposphere.

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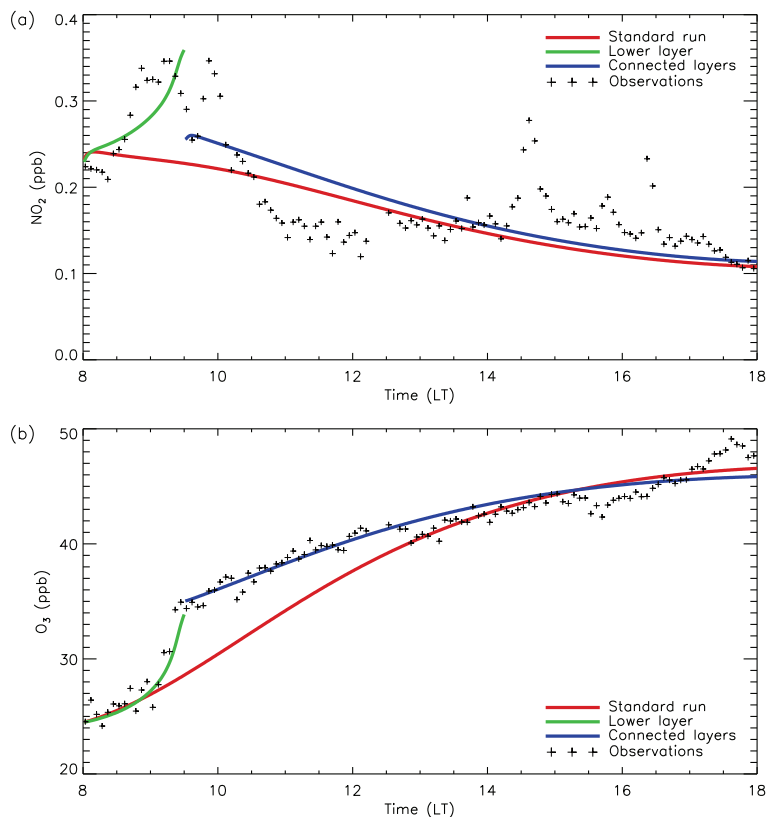


Fig. 8. Comparison between the observations and MXL model results for the **(a)** NO₂ and **(b)** O₃ mixing ratios during 6 August 2010. The numerical experiments are the standard case, the case for the lower shallow boundary layer until 9:30 LT and the case where a residual layer and the lower shallow boundary layer are connected and become one mixed layer from 9:30 LT onward. The latter two cases correspond to situation **(1)** and situation **(3)** in Fig. 2, respectively.

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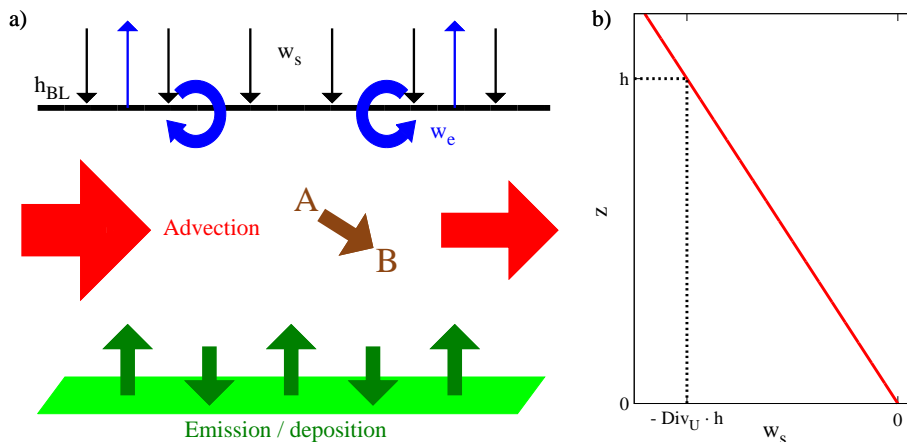


Fig. 9. (a) Schematic overview of the different terms that affect the evolution of scalars and the boundary layer height. Shown are the impact of emissions and deposition near the surface, advection, entrainment, additional sources and sinks (e.g. due to chemical conversion) and subsidence. The dependence of the subsidence velocity on height is presented in (b).