

We thank referee #3 for his valuable comments and suggestions. We followed them as explained below.

The reviewers comments are repeated in **bold letters**, our replies are given in *italics*, and text modified or added to the manuscript is given in blue.

GENERAL REMARKS

**The paper would be worthwhile for publication in ACP if the following major remarks were properly taken into account, in particular:
a sound argumentation on the choice of several parameters (in particular fire intensity),
a better description on how optical properties and aerosol-cloud interactions are calculated in the model.**

Both points are captured in detail in the following.

MAJOR REMARKS

Section 2.1, page 5, lines 13 – 20. The parameters used in this study to obtain the lower and upper bounds of the plume height need to be much better justified, in the context of available fire studies. When reading the paper, one could think that the two upper and lower values given for fire intensity (30 and 80 kw/m²) represent a common range of observed values. But then these values are not really used in a statistical sense, but rather in a deterministic way to calculate lower and upper plume heights for a given fire. Isn't there a conceptual mismatch. Also the values chosen for the limiting vertical velocity and default fire size need justification. For all values, how would altering them with respect to their estimated uncertainty ranges alter the results of this paper. Some sensitivity tests would be welcome here.

The methodology for using a range of values for heat flux and sensitivity tests made with the plume rise model is described in Freitas et al. (2007). [The application of a range of heat flux is justified not only by the variability associated with the vegetation condition, which is not known, but also by the own dynamic variation during the combustion process. Besides, this range is also applied in a statistical sense since the net emission in the 3-D atmospheric transport model might be associated not with a unique fire but a set of sub-grid scale fires all burning inside the same model grid box. Using the fire radiative power \(FRP\) to estimate the buoyancy flux does not help to eliminate the use of the prescribed range of the heat flux, since there is still a substantial uncertainty in converting FRP to the convective energy, which has been widely described in the literature \(Wooster et al., 2005, Val Martin et al., 2012, Paugam et al., 2015\). Moreover, the uncertainty in the FRP retrieval by sensors on-board of satellites is also high.](#)

Section 2.4, page 7: This section is difficult to read, because the aim of the argumentation is not clear from the beginning on. The last sentence, that the authors were unable to perform Mie calculations for this study, and thus took values for diesel soot instead of wood soot should be put right in the beginning of the section. Potential implications of this approximation should be discussed all along the paper, in particular in section 3.5 (radiative effects).

We have completely rewritten this section following the reviewers' suggestions. Unfortunately, our original statement regarding the single scattering albedo of biomass burning aerosol and pure diesel soot particles was misleading. In COSMO-ART soot particles are subject to aging during the transport process and therefore also the single scattering albedo of the simulated aerosol population increases. [Calculating the radiative effect of biomass burning and other aerosol types requires the optical properties extinction coefficient, single scattering albedo, and asymmetry parameter of the aerosol particles at each grid point and each time step. These optical properties depend on the refractive](#)

index of the individual compounds, the chemical composition of the particles, their shape, and their size distribution. The refractive index and therefore the optical properties depend on the wavelength.

Insoluble light absorbing particles like soot can be covered by a soluble shell due to physical (coagulation, condensation), and photo-chemical ageing. This increases their mass absorption efficiency (Riemer et al., 2003; Saleh et al., 2014; Bond et al., 2013). That effect needs to be accounted for in fully online-coupled model systems like COSMO-ART. Mie-calculations are the adequate method to determine the optical properties from given size distributions and their chemical composition (Bohren and Huffman, 2004). These calculations are very time consuming and therefore it is not possible to perform them at each grid point and at each time step. Instead, we have developed a parameterization as described in Vogel et al. (2009). This parameterization is based on simulated aerosol distributions and detailed Mie-calculations ending in mass specific values of the extinction coefficient, single scattering albedo and asymmetry parameter. Moreover, this parameterization takes into account the physical and chemical ageing of soot particles (Riemer et al., 2004; Vogel et al., 2009). Values are delivered for the wavelength bands of the radiation scheme used in COSMO-ART (Ritter and Geleyn, 1992). Fundamental input data for the Mie-calculations are the wavelength dependent refractive indices for the individual compounds. Here, we are using data of detailed measurements performed in the AIDA (Aerosol Interaction and Dynamics in the Atmosphere) chamber (Schnaiter et al., 2003). The disadvantage of this data is that it was obtained for pure diesel soot. But its advantage is the high spectral resolution of the data which is not the case for other lab studies. A comparison of this fundamental input data with data obtained for biomass burning aerosol is difficult for several reasons. Recent studies ended up with bulk data for mostly aged particles or with mass specific values for extinction and absorption coefficients. Consequently, quite different values were found depending on the specific burning conditions and particle compositions. In many cases values were gained for a single wavelength. For that reason it is hard to quantify the errors due to the calculation of the optical properties within COSMO-ART.

Following our parameterization we get a value for the mass extinction efficiency of $9.0 \text{ m}^2 \text{ g}^{-1}$ for the spectral range 0.25 - 0.7 μm and for pure soot particles. For the soot containing Aitken mode we get a value of $5.0 \text{ m}^2 \text{ g}^{-1}$, and for the soot containing accumulation mode a value of $4.0 \text{ m}^2 \text{ g}^{-1}$. Laser measurements at a wavelength of 0.632 μm suggest a value of $7.8 \text{ m}^2 \text{ g}^{-1}$ for soot with wood origin (Colbeck et al., 1997).

Levin et al. (2010) carried out measurements with biomass burning aerosol of different chemical composition. The geometric mean diameters ranged from 0.2 - 0.57 μm . For those particles they found refractive indices ranging from 1.55 - 1.80 for the real part and 0.01 - 0.50 for the imaginary part. They obtained dry mass extinction efficiency ranging from 1.64 - 6.64 $\text{m}^2 \text{ g}^{-1}$ at a wavelength of 0.532 μm . Hungershoefer et al. (2008) found mass extinction efficiencies in the order of $9.0 \text{ m}^2 \text{ g}^{-1}$ for savanna grass and African hardwood.

From these numbers we would conclude that the optical properties we are using are within the range of literature data.

Fire aerosol is also constituted of organic aerosol. Which optical properties are adopted for organic aerosol? Is internal or external mixing assumed for different fire aerosol components? This should be stated. Only one reference for one wavelength is given for the single scattering albedo of diesel and wood. I guess that there are much more results available in literature. Please synthesize. Optical parameters of soot have been shown to change with plume age (for example review of Bond et al., 2013). This effect is not considered in the present study. This point should at least be discussed. Please also discuss, how specific information on size distribution would ideally be used for Mie calculations, and how this was handled in the present study. Again, what is the expected error?

This comment is addressed within the new version of section 2.4. Regarding the aging process we were not clear enough within the manuscript. In comparison to many other models it is a great advantage of COSMO-ART that it treats the aging of soot particles explicitly. Soot is treated as an

external mixture after its emissions and is then transferred by coagulation and chemical aging into an internal mixture (Riemer et al., 2003; Riemer et al., 2004).

Additional section 2.5: Please describe, how aerosol microphysics interactions are treated in the model, which processes and parameterizations are included? This is crucial for enabling the reader to understand results presented in Section 3.5 (Aerosol radiative impact).

The general model description was extended by specifications for the aerosol radiation interactions and aerosol cloud interactions.

The simulations are conducted using the comprehensive online-coupled model system COSMO-ART (Consortium for Smallscale Modelling - Aerosols and Reactive Trace gases, Vogel et al., 2009). This system is based on the operational weather forecast model COSMO (Baldauf et al., 2011). COSMO-ART includes a comprehensive chemistry module to describe the gaseous composition of the atmosphere and secondary aerosol formation, and it allows for feedback of the simulated aerosol particles with radiation, cloud formation, and precipitation (Stanelle et al., 2010; Knote et al., 2011; Bangert et al., 2012; Lundgren et al., 2013; Athanasopoulou et al., 2014; Rieger et al., 2014; Vogel et al., 2014). The size distribution of aerosol within COSMO-ART is approximated by log-normal distributions. In Table 1, all required modes with their initial median diameters, standard deviations and chemical compositions are presented. The standard deviation is maintained constant while the median diameter of the aerosol changes during transport. Chemical reactions are calculated with RADMKa (Regional Acid Deposition Model Version Karlsruhe, Vogel et al., 2009) which is based on RADM2 (Regional Acid Deposition Model, Stockwell et al. 1990). The formation of secondary organic aerosol is calculated by a VBS approach (volatility basis set, Athanasopoulou et al. 2012). COSMO-ART explicitly treats the aging of soot particles transferring them from external to internal mixtures as described in Riemer et al. (2003). The radiative fluxes are calculated with the GRAALS radiation scheme (Ritter and Geleyn, 1992). Preliminary Mie-calculations have been performed for the initial aerosol particle size distributions and their chemical composition to obtain mass specific values for the extinction coefficient, single scattering albedo, and asymmetry parameter. These coefficients also depend on wavelength. To consider the optical properties of the current aerosol distribution the mass specific parameters obtained by the Mie-calculation are weighted with the mass fraction of the chemical components. Within COSMO-ART a full two-moment cloud microphysics scheme (Seifert and Beheng, 2006) is used. Aerosol activation is considered according to Fountoukis and Nenes (2005). Ice nucleation is based on the parameterization by Barahona and Nenes (2009a, b).

Section 3.4: The arguments given for stating that the VARHEIGHT simulation is the best are to some extent convincing. Nevertheless, the given data set is quite restricted, are there more observations available? For instance in-situ PM measurements at surface sites? MODIS or POLDER AOD fields? Is it possible to put the discussion on a more quantitative basis (for example by calculation of correlation coefficients between simulations and observations?)

We added a comparison with MODIS AOD fields (see comment below).

It should be mentioned while discussing results in section 3.4, that differences between simulations and observations could be due also to errors in fire intensity and emissions. In how far do such errors prohibit from drawing conclusions on the different plume rise schemes.

We added:

Note that if errors are made in the estimate of fire intensity and emissions this will influence the concentration in all simulations, while the plume height is only affected in VARHEIGHT and EMISSCYCLE.

Overall, section 3.4 is quite difficult to follow, may be it is possible to simplify, and not give all numbers. Those could be grouped together in a table.

We shortened this paragraph, and modified the corresponding figure.

The most prominent features of the observed smoke distribution are marked with dark green circles. Circle A indicates smoke observed by CALIOP between 6 and 7.5 km altitude. This feature is well represented by the simulations 7500M and EMISSCYCLE, moderately represented in VARHEIGHT and 800M fails at this point. Circle B refers to smoke within the lowest 3.5 km. In all simulations the smoke is located a little lower at this position but each of them showing distinct patterns in each case. Circle C and the descending line represent the skewness of the smoke layer between 56 and 50° N. The decline seems to be stronger in the simulations than in the observations. The height is matched by simulation VARHEIGHT and 800M. In EMISSCYCLE the height is slightly overestimated and in 7500M the height is remarkably overestimated.

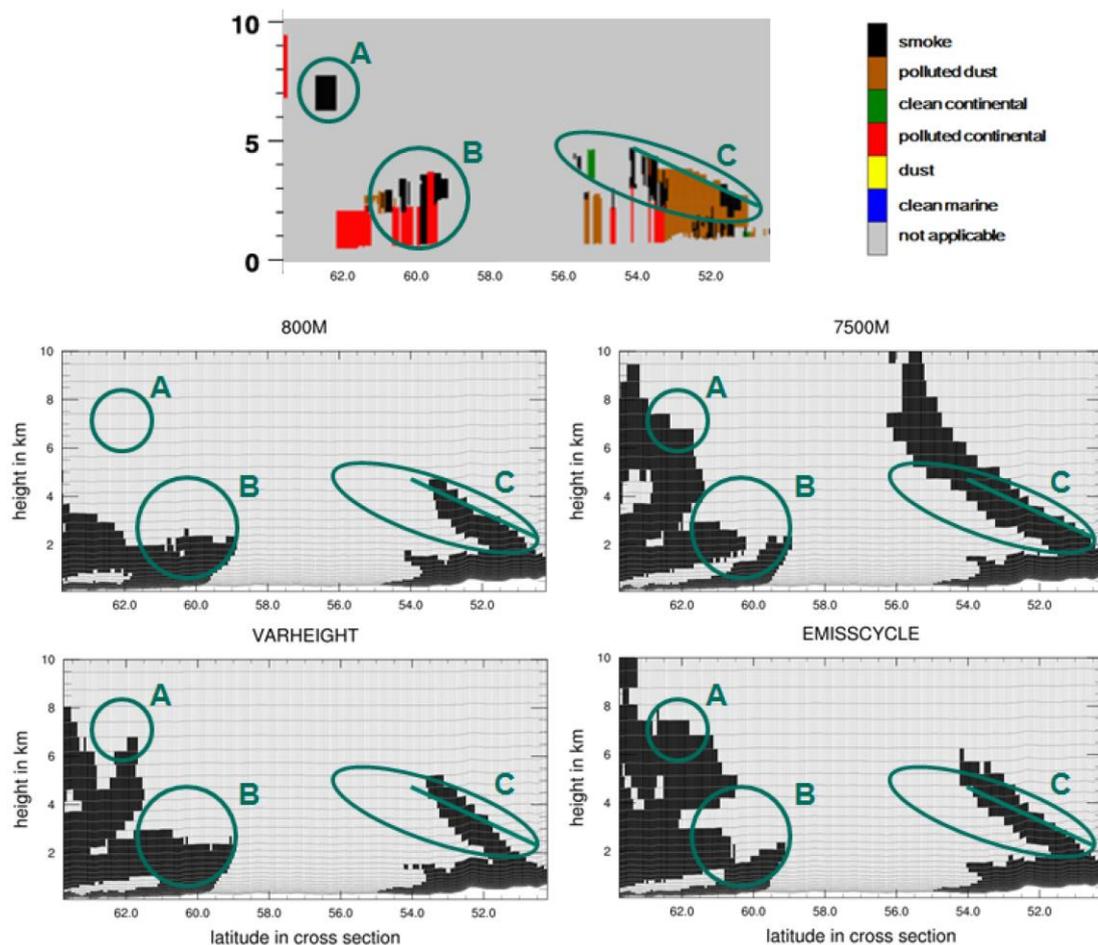


Figure 9. Cross section of aerosol subtypes of CALIPSO overpass at around 9:20 UTC 16 July 2010 (a). The black colour coding denotes the presence of smoke; brown, green, and red represents polluted dust, clean continental, and polluted continental, respectively. Cross section along the same CALIPSO track for simulations 800M (b), 7500M (c), VARHEIGHT (d) and EMISSCYCLE (e), here only soot concentrations greater than $0.01 \mu\text{g m}^{-3}$ are displayed.

Section 3.5 would be strengthened, if simulated effects on short wave radiation, temperature and cloud cover could be substantiated by observations, for the given case study. This should be possible from meteorological in situ and satellite observations. Without observations, this section remains rather speculative.

We now included station measurements of the short wave radiation at Fort Smith:

Observations of the global solar radiation at Fort Smith (60.01° N, 111.57° W, Meteomanz.com) do support these simulations. On 15 July 2010 at 6 UTC the station reports 1115 J cm⁻² during the last 24 hours. The simulation VARHEIGHT which includes the fire emissions yields 1029 J cm⁻² for the same 24 hour period, whilst the simulation NOFIRE results in 2222 J cm⁻². This is a typical value for cloudless, smoke-free days. For example on 11 July 2010 a value of 2168 J cm⁻² was reported at that station.

In addition a satellite retrieval of AOD is added, this is specified in more detail later on.

MINOR REMARKS

Page 3, lines 7-9: Is this rapid transport to Europe due to prior vertical lifting into the upper troposphere with stronger winds. Please make this link clear in the revised text.

At this point we added: This is due to lifting into high altitudes by pyro-convection prior to horizontal advection over the North Atlantic Ocean.

Page 3, lines 19-23: are these arguments valid for specific cases or are they more general, please make this clear.

We only want to refer to their observations without any assumptions on generality.

We added: in their case

Page 4, lines 11-19: please better argue, why this study is new with respect to older work.

To the best of our knowledge we are the first to investigate the effect of biomass burning aerosol on temperature and dynamics with an online-coupled modelling system on synoptic time scales with an explicit treatment of the aging of soot in combination with a plume rise model.

Page 4, model description: Is secondary aerosol formation from biomass burning emissions included in the model? This process is for example shown to be important for Russian fires in summer 2010 (Konovalov et al., 2015).

We have extended section 2 to give a more clear and comprehensive description of COSMO-ART regarding the aerosol treatment. The VBS scheme included in COSMO-ART is described in Athanasopoulou et al. (2012).

Page 5, Section 2.1: is lateral detrainment in the convective fire plume is apparently not considered?

In our version of the plume rise model only entrainment of environmental air into the plume is considered. In a more recent version of the plume rise model detrainment was included (Paugam et al., 2015).

Page 7, line 18: 'in sufficient agreement' agreement with what?

This sentence has been removed from the text.

Page 11, line 32: A median mass diameter above 1µg/m3 seems large to me. It is for instance larger than the accumulation mode in which most mass of continental aged pollution aerosol is concentrated. Is there an explanation, why this is different for fire aerosol.

Since mass size distributions for biomass burning aerosol seem to be very rare, we decided to replace it by a comparison with a number size distribution from a laboratory measurement.

The simulated number distributions for Fort Smith (60.01° N, 111.57° W; Fig. 3), a location in the fire (61.30° N, 110.45° W), and a location in the vicinity of the fire (58.12° N, 106.51° W) near the surface

on 15 July 2010 at 18:00 UTC are shown in Fig. 11. Unfortunately, we have no in-situ characterization of the aerosol particles. Instead we compare the model results with the size distribution measured during a small-scale laboratory experiment performed by Hungershoefer et al. (2008). For their experiment savanna grass and African hardwood were burnt in a smoke chamber in order to characterize the optical properties of biomass burning aerosol. At Fort Smith the simulated number concentration is about three orders of magnitude smaller than in the laboratory measurement, while the median diameter is about $0.1 \mu\text{m}$ in both cases. At the fire the simulated number concentration is comparable to the measurement but the simulated median diameter of $0.04 \mu\text{m}$ is smaller than in the measurement. Close to the fire the concentration gets smaller and the median diameter bigger than at the fire. The diameter is still smaller and the concentration still higher than in Fort Smith which is located further away from the fire. The aging process clearly arises out of the increasing median diameter with distance to the fire. Especially close to the fire the course of measurement and simulation show reasonable agreement. A smaller number concentration can be expected due to dispersion of fresh air outside a laboratory.

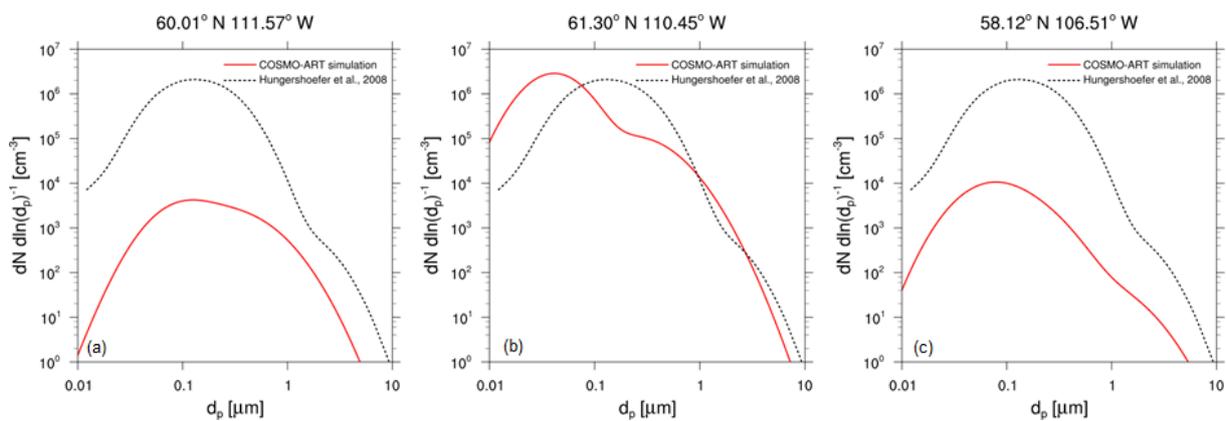


Figure 11. Number size distribution (a) at 60.01° N , 111.57° W (Fort Smith), (b) 61.30° N , 110.45° W (at fire), and (c) 58.12° N , 106.51° W (close to fire) at the surface on 15 July 2010 at 18:00 UTC for VARHEIGHT (red line) and as comparison measurements from an experimental fire (dotted black line) performed by Hungershoefer et al. (2008).

Tables:

It would be worthwhile to add a table with emission factors for different model species.

We added a new table which contains the emitted species and their assignment to the existent COSMO-ART classes. Furthermore, we added the following sentence.

The species are listed in Table 2 together with their assignments and individual weightings, where necessary.

Table 2. Emitted gaseous and particulate species derived from GFASv1.1

original notation	COSMO-ART class
Carbon Monoxide	CO
Nitrogen Oxides $\text{NO}_x * 0.9$	NO
Nitrogen Oxides $\text{NO}_x * 0.1$	NO2
Sulfur Dioxide	SO2
Ammonia (NH_3)	NH3
Ethane (C_2H_6)	ETH (Ethan)
Methanol (CH_3OH)	HC3 (C_3 to C_5 Alkanes)
Ethanol ($\text{C}_2\text{H}_5\text{OH}$)	HC3 (C_3 to C_5 Alkanes)
Propane (C_3H_8)	HC3 (C_3 to C_5 Alkanes)
Butanes (C_4H_{10})	HC3 (C_3 to C_5 Alkanes)
Pentanes (C_5H_{12})	HC5 (C_6 to C_8 Alkanes)
Hexanes (C_6H_{14})	HC5 (C_6 to C_8 Alkanes)
Heptane (C_7H_{16})	HC8 (higher Alkanes)
Ethene (C_2H_4)	OL2 (Ethene)
Propene (C_3H_6)	OLT (terminal Alkenes)
Butenes (C_4H_8)	OLT (terminal Alkenes)
Octene (C_8H_{16})	OLT (terminal Alkenes)
Pentenes (C_5H_{10})	OLI (internal Alkenes)
Hexene (C_6H_{12})	OLI (internal Alkenes)
Isoprene (C_5H_8)	ISO (Isoprene)
Terpenes (C_5H_8) _n	API (Terpenes)
Toluene (C_7H_8)	TOL (Toluene)
Benzene (C_6H_6)	TOL (Toluene)
Xylene (C_8H_{10})	XYL (Xylene)
Formaldehyde (CH_2O)	HCHO
Acetaldehyde ($\text{C}_2\text{H}_4\text{O}$)	ALD (Acetaldehyde)
Acetone ($\text{C}_3\text{H}_6\text{O}$)	KET (Ketones)
Black Carbon	s (pure soot mode)
Organic Carbon * 0.1	if (Aitken mode particles, soot free)
Organic Carbon * 0.9	jf (Accumulation mode particles, soot free)

Figures:

Figure 2: please specify the Figure legend, for which parameter the diurnal cycle is shown?

We changed the figure legend. It now reads as:

The course of the diurnal cycle assumed for fires in boreal forests. In the individual simulations this diurnal cycle is overlaid on the daily values of fire size, fire intensity and emission strength.

Figure 4: What is the meaning of the red points? I guess the smoke area is in grey, while clouds are white. To be completely clear, this could be mentioned in the legend.

We added:

The red dots denote the fire locations. The grey structures state the distribution of smoke.

How does observed smoke region compare to that simulated with different plume height options. Does such a comparison allow state on benefits of different plume height treatments?

In order to address this point we added an additional figure and the following text.

To evaluate the horizontal diffusion of the plume the simulated AOD is compared with AOD satellite retrievals, both at 550 nm. In the top of Fig. 8 observations made by MODIS on-board Terra and retrieved with the dark target algorithm are displayed time averaged over 14 and 15 July 2010. Below the AOD averaged over the four overpass times of Terra satellite are shown for the different simulations. The observed maximum of over 3.5 is located around 57.5° N, 112.5° W. From there the increased AOD is spread towards north-east and south-east. In all simulations the maximum is located further in the east than in the satellite retrieval. The pattern of AOD differs between all simulations in its width, shape, and strength. The southern extension of the plume reaching 50° N, 105° W is best represented by the simulations VARHEIGHT and 800M. Due to the coarse resolution of the satellite retrieval it is not possible to determine the overall best match.

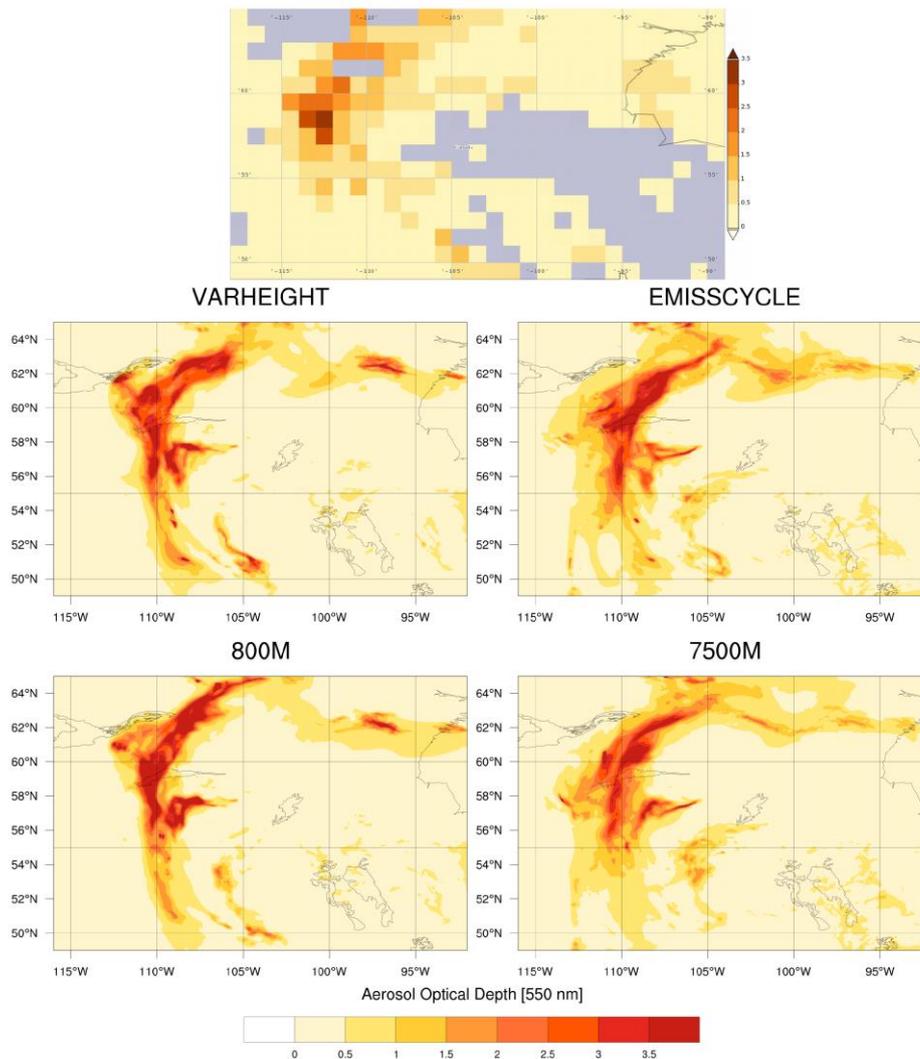


Figure 8. AOD at 550 nm averaged over 14-15 July 2010. Top: Satellite retrieval from MODIS on-board Terra, below: Simulations VARHEIGHT, EMISSIONCYCLE, 800M, and 7500M.

Figure 5: It is difficult to make the “geographical” link between figures 4 and 5. In figure 5, please indicate latitudes and longitudes, or make appear the domain of fig. 4 in fig. 5.

The simulation domain shown in Fig. 3 and Fig. 4 is now indicated in Fig. 5. We added to the figure legend:

The edges of the simulation domain are indicated with blue triangles.

Figure 7: How are colors attributed, it is not very quantitative?

No, it is not quantitative. The aerosol subtypes are determined with an associated aerosol lidar ratio at 532 nm and 1064 nm. With this only a classification of the aerosol is treated.

This is mentioned in the main text, please recall it in the figure legend.

We added:

The black colour coding denotes the presence of smoke; brown, green, and red represents polluted dust, clean continental, and polluted continental, respectively.

Figure 10: Aren't there any observations of short wave radiation available in the modelling domain?

We now included station measurements of the short wave radiation at Fort Smith:

Observations of the global solar radiation at Fort Smith (60.01° N, 111.57° W, Meteomanz.com) do support these simulations. On 15 July 2010 at 6 UTC the station reports 1115 J cm⁻² during the last 24 hours. The simulation VARHEIGHT which includes the fire emissions yields 1029 J cm⁻² for the same 24 hour period, whilst the simulation NOFIRE results in 2222 J cm⁻². In turn this value is a normal finding for cloudless, smoke-free days, e.g. on 11 July 2010 a value of 2168 J cm⁻² was reported at that station.

Is figure 10 contained in Figure 13, or is it different. It could be justified, but please indicate it.

Yes, simulation VARHEIGHT is depicted in both figures. We put these two figures into one.

TECHNICAL, EDITING REMARKS

Page 2, line 7: 'they developed' -> 'Konovalov et al.' or 'the authors'

Done

Page 3, line 18: 'Another simulation....' In the same study/reference?

We changed another to [the](#). This simulation was performed in the same study.

Page 3, line 18: 'the same' Which ?

The source height of 800 m was meant. We added: [of 800 m](#)

Page 6, line 18: 'the wind speed in the boundary layer is usually higher....' Add 'usually'

Done

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