Response to anonymous reviewer’s #1 comments.

The authors thank the reviewer for their constructive comments. Our responses to anonymous reviewer's #1 comments are detailed below. Reviewer’s comments are in italics and our responses in standard font.

General Comments:

Section 2.3 (Plume Dispersion Modelling) is not well described. 1) Was the same particle number used for the two fire types (boreal and temperate)? Is this reasonable? Are there references to support this approach? 2) What are the two different rates that are used for day and night emissions? Are there references to support different day/night rates? 3) When no fires were detected, why set the count to a minimum positive value instead of zero? Is this to account for undetected fires? What support is there for this approach? Please rewrite this section for clarity.

The authors apologise for the lack of clarity in the section. The section has been rewritten clarifying the steps undertaken to perform dispersion simulations, and addressing the specific questions raised in this comment. The answers (numbered in the reviewers comment) are listed below.

1) HYSPLIT particle number emitted per hour per fire detection was the same for both biomes. This was done for several reasons. Firstly, total particle number is not directly linked to particulate emission estimates. For example, if a grid cell has AOT value of 1, and 100 HYSPLIT particles are located within the cell during the satellite overpass, 80 of which were emitted two diurnal cycles ago, and 20 during the previous diurnal cycle, the grid cells AOT is split accordingly between the emission periods. Equivalently, if there are any particles emitted from different fire events, grid cells AOT is divided both between different emission periods and different fire events. The estimates for the two periods would not be different if there were more or less particles within the cell, its the relative ratios which matter. Secondly, the aim of the manuscript was to provide independent estimates therefore we tried to avoid using existing emission factors or emission coefficients (top-down emission estimates per unit of fire radiative energy). Notably, even if emission rates are indeed different for boreal and temperate events, the assumed identical particle numbers in the analysis would not have influenced the results significantly. This is due to the fact that mixing of the plumes from different biomes was minimal.
2) The emitted particle number per hour and fire detection were identical for daytime and nigh-time periods.
3) The count was set to minimum non-zero value to avoid total shutdown of emissions for a time period for which no MODIS fire detections were obtained, most likely due to cloud cover. This is an unlikely scenario for the long duration burning episodes presented in the manuscript.

The updated section:

Smoke transport for the selected fire events was simulated with the HYSPLIT model (Draxler et al., 2003). Plume dispersion from a source location was represented by the motion of a large number of discrete particles moved by the wind field with mean and random components. Global Data Assimilation System (GDAS) meteorological archive data was employed to drive the model.

For each day of burning, particles were continuously released into the model domain from the locations of the individual active fire detections within the fire event. In order to represent fire diurnal cycle, different MODIS active fire observations were used to release particles for two 12 hour intervals representing day and night emissions 09:00 to 21:00 and 21:00 to 09:00 local time respectively. Emission source number and locations for daytime periods were determined from the highest number of fire detections observed during a single either Terra of Aqua daytime overpass with 10.30 and 13.30 equatorial crossing time. Similarly, emitted particle source numbers for the night periods were determined by the largest burning extent observed during one of the night-time overpasses with 22.30 and 1.30 equatorial crossing times. Notably, the Terra overpass at 22.30 in high latitudes makes observations of regions where local time is earlier than 21:00. In this study, however, all fires detected during this overpass were classed as night-time observations. If no valid observations were available for some of the time intervals, the count and fire pixel locations were set to a~minimum non-zero value estimated for the burning episode from all daytime or night-time observations. This was done to avoid total temporary shut-down of the emissions, which is an unlikely scenario for a long duration burning episodes. Every hour, 20 particles were released for each fire pixel. As a result, emitted particle number for a burning episode was determined by the number of active fire pixels observed during a given time period.

Particles were uniformly distributed between the surface and the top altitude of the planetary boundary layer as given in GDAS archive. Satellite based plume height estimates (Val Martin et al., 2010, Peterson et al., 2014) indicate that in up to 80% of the events analysed, injection heights were limited to the planetary boundary layer. While confinement of the emissions to the mixing layer underestimates injection height for the most energetic burning episodes, such configuration should nonetheless represent the majority of burning episodes.

Throughout the simulations, modelled particle positions, their age and source burning event identifier were recorded each day at local solar noon. The generated point clouds were later used to compare against Terra and Aqua Aerosol Optical Thickness (AOT) observations. "

Water Content Retrieval. How do aerosol water fractions estimated in this work compare to aerosol water content that would be estimated using representative hygroscopic growth factors for representative relatively humidities?

The inferred median water volume fractions would equate to geometric hygroscopic growth factors (gHGF) of 1.05 and 1.24 for boreal and temperate plumes respectively. Such factors suggest that boreal plumes belong to "nearly-hydrophobic'' group (gHGF 1.0-1.11) while temperate plumes fall into ''less-hygroscopic'' category (gHGF range 1.11–1.33) as suggested in Swietlicky et al., 2008 review of measured growth factors. Note that the measured growth factors were recorded at 90% relative humidity, while water fractions inferred in our study were obtained at ambient relative humidities. In result, a direct comparison is not very meaningful, but our numbers do seem to fit reasonably well. Smoke aerosols are most often classified as less-hygroscopic with gHGFs of 1.11-1.33. We have added a short discussion:

"These estimates compare favourably to measured factors for biomass burning smoke (Swietlicky et al., 2008), indicating nearly-hydrophobic particles for boreal plumes, while temperate plumes could be classed as less-hygroscopic. Notably, measured geometric hygroscopic growth factors are reported at 90% relative humidity. In contrast, water volume fractions inferred in this study are representative of ambient humidity levels, and as a result direct comparison is not very meaningful.

The authors use "emission coefficients" in the text. Are these distinct from emission factors? If not, then rather use emission factors, as this is common terminology. If so, then please clarify the distinction in the text.

The term "emission coefficients" is used to contrast the top-down particulate emission estimates per unit of FRP with measured emission factors which are obtained for unit of fuel burned. Such nomenclature was used in other top-down studies (Kaiser et al., 2012; Ichoku and Elison 2014). We have rephrased the paragraph introducing the concept:

“A~top-down global gridded Fire Energetics and Emissions Research (FEERv1) (Ichoku et al., 2014) product is based on collocated satellite FRP and AOT observations. Inferred total particular matter emissions rates are linked to observed FRP. The estimated TPM emission coefficients allow direct conversion from time integrated FRP to emitted particulate matter without invoking the emissions factors.”

Specific Comments:

Abstract: FRP is used in the Abstract (and Introduction), but the acronym is only defined on page 10.

We have replaced the acronym in abstract with “fire radiative power”, and added the definition to the first instance in the text.

Abstract, line 20: Is “low bias” meant to be “negative bias”?

Indeed, “negative bias” was the intended wording, changed accordingly.

Page 2, line 19: Is the 3.4 correction factor applied to address an underestimate or overestimate in emissions? Please be specific.

The correction factor was needed to address underestimation in emissions. The ambiguous statement has been changed to “enhancement factor”.

Page 2, lines 32-34: This sentence is confusing. Please reword for clarity. Currently it reads that average EFs conceal the lack of spatial and temporal representativeness. Is this what the authors mean to say?

The sentence has been rephrased to “Average EFs for different biomes are based on small sample numbers for some areas, and conceal large variability in individual measurements…”

Page 3, lines 2-3: Isn’t the approach in this study also susceptible to AOT retrieval errors and uncertainties in smoke particle properties?

We perhaps didn’t make this clear enough, but the statement was “top-down aerosol inversions are affected by AOT retrieval error and large uncertainties in assumed smoke particle properties”. The approach taken by this study is “top-down” by definition, therefore the statement was directed at our and other similar studies. To make it clear that it applies to any top-down method including this study, we have replaced “top-down aerosol inversions” with “top-down approaches”.

“Page 3, line 24: Please provide an appropriate reference or website for the MCD14ML data.

Reference (Giglio et al., 2006) has been added.

Page 4, lines 2-3: The approach used to identify fires is confusing. Are the “any pixels” pixels that include an active fire? Should there be consecutive active fire pixels within a 150 km radius? Please clarify in the text.

“any pixels” was used meaning any MODIS fire detections. This whole section has been rewritten for clarity as been requested by the other reviewer:

Large and long-lived fire events, likely strong emission sources, were identified and selected for the analysis. Burning episodes larger than 100km^2 are not numerous, but account for more than 80%

of total burned area in boreal North America (Stocks 2002, Kasischke 2002), and are a dominant mode of burning in parts of temperate regions as well (Strauss et al., 1989). In order to identify such events, individual MODIS active fire detections were agglomerated into large wildfire events by performing two step spatial-temporal clustering. First, any MODIS fire detections located closer than 10km in space and 24 hours in time were grouped together. Single detections not assigned to any of the formed clusters were removed from further analysis. The clusters were then filtered by selecting events with (i) spatial bounding box containing all fire detections belonging to the cluster larger than 100km^2 and (ii) duration longer than 7 days. The duration was determined by the time span between the first and the last MODIS active fire detection belonging to the cluster. The burning was considered uninterrupted if the largest temporal interval between subsequent MODIS fire observations was less than 24 hours. During the second step of clustering, any of the selected events active at the same time and located closer than 150km were grouped into large burning episodes, assigning a unique source label. These events were classified into boreal and temperate fires using the dominant emission source given in the GFEDv4 inventory for areas and periods when the events were active.

Page 5, lines 10-11: The change in resolution from nadir to the swath edges is true for the native resolution of the instrument, but the AOT product is at a nominal resolution of 10 km x 10 km.

It is our understanding that Collection 5 MODIS dark target algorithm uses blocks of 20 x 20 500m MODIS pixels while deep blue algorithm employs block 10 x 10 of 1 km MODIS pixels (Levy et al., 2013; Sayer et al., 2015). The block size is fixed across the swath, and hence the footprint increases proportionally with individual pixel size. The original statement in the manuscript “pixel size increases twice at the edge of the swath” was indeed incorrect as pointed out by the other reviewer. In fact, AOT pixel footprint increases approximately 9 times.

Page 5, line 12: More appropriate is the MODIS AOT uncertainty for scenes with aerosols from boreal and temperate fires. Please either estimate the error by comparing MODIS and AERONET AOT or provide a value reported in the literature.

We have added the regional analysis of Collection 5 MODIS AOT retrieval uncertainties (Hyer et al., 2011) to the paragraph:

“A regional MODIS M{*}D04_L2 AOT product validation (Hyer et al., 2011) indicates that performance varies greatly within North America. The study found that for 0.2<AOT<1.4 conditions, root mean squared error varies from 0.01+0.51xAOT in arid Western America where retrieval is hindered by bright surfaces, to 0.01+0.31xAOT in boreal forests and 0.3+0.12xAOT in Eastern USA. The study reported positive bias in MODIS AOT for some locations, in particular for retrievals at extremely high aerosol loadings.”

Figure 3: What is the mass concentration of individual aerosol components (inorganic, organic, black carbon) from boreal and temperate fires estimated in this study?

While we did estimate volume fractions of black carbon and a mixture of organic and inorganic compounds represented by n values close to 1.53 when retrieving water content, a decision was taken not to present them in this manuscript. We believe that the discussion on different components should be left out of this manuscript which is focused on TPM estimates. In any case, the retrieved median volume fractions of black carbon were relatively low and hardly different when comparing the two categories, 0.01 and 0.009 for boreal and temperate plume observations respectively. Notably, we did not retrieve absorbing organic carbon or “brown carbon” fractions by utilising its wavelength dependent absorption. Organic carbon was represented as part of the third component encompassing organic and inorganic compounds.

Page 6, lines 6-7: This sentence is unclear. Is this merely a scaling to convert AOT to fire-emitted particle number?

The section has been updated clarifying the steps used when attributing AOT. Bellow is the relevant paragraph:

"If a mixture of particles was found within a cell indicating that multiple fires and multiple emission periods contributed towards the grid cell AOT, the attribution was performed by apportioning a~grid cell's fire-emitted AOT in proportion to the numbers of modelled particles released during the emission periods and with origin found within the grid cell. For example, if a grid cell had AOT value of 1, and 100 HYSPLIT particles were located within the cell during the satellite overpass, 80 of which were emitted two diurnal cycles ago, and 20 during the previous diurnal cycle, the grid cell AOT was split accordingly between the emission periods. Panels K and L in figure 1 illustrate partitioning of total plume AOT to two different emission periods. Similarly, if there were any particles emitted from different fire events, grid cells AOT was divided both between different emission periods and different fire events."

Table 1: Please change density to the Greek letter "rho" and enclose the units for density in square brackets for clarity.

Thank you for noting this, the symbol was changed accordingly.

Page 7, line 19: Why list both "organic carbon" and "organic matter"?

Thank you for pointing this out. The redundant phrase has been removed.

Page 8, lines 19-20: Incorrect in-text citation format for Ichoku and Ellison (2014). Please fix.

The citation has been fixed.

Page 10, lines 27-28: How different is median FRP for the two fire classifications if normalized to burned area or mass of biome burned?

We do agree that such a comparison would have been interesting. However, we did not employ area burned datasets in the study, and did not perform a comparison with biomass burned estimates. As a result, it is not feasible to include this information at this stage. In any case, the comparison of per-pixel FRP values is only a sideline to this study, and we did not place much weight on it, but felt the need to report it.

Page 10, line 33: Would smouldering fires be detected as part of the large wildfire events that are isolated in this work?

The authors are not aware of a method to directly detect smouldering combustion by remote sensing means. Median FRP values alone certainly do not provide enough information. However, smouldering combustion has been reported to be more important during night (Reid et al 2005); and that smoke from smouldering combustion can be lofted and entrained into main plumes by convection (Urbanski 2013). The statement is therefore merely an interpretation based on literature, not something which can be confirmed by the data employed.
To avoid any possible confusion, We have removed the statement linking lower FRP and night time burning to smouldering combustion.

Page 11, line 15: What is "(4)" referring to? Is this Figure 4?

The reference to figure 4 has been corrected.

Page 11, line 17: Are the median water volume values fraction or percent? Fraction is stated, but the "%" symbol is used (page 12, line 1).

We apologise for this error. The median volume fractions were reported, not percentages. The symbol has been removed.

Figure 5: This figure is out of sequence in the text. It is referred to in the text before Figure 4. Please fix

Figure reference order has been corrected.

Page 14, line 17: Is 80% relative humidity?

Yes, relative humidity was meant. Added to the text.