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Abstract

A new black carbon (BC) removal scheme with increased hydroscopicity is implemented in the Coupled Large scale Aerosol Simulator for Studies In Climate (CLASSIC) and tested against the standard scheme using a present day climate simulation. The lifetime of black carbon is reduced from 16.2 days to 6.0 days when using the new scheme. This new lifetime is consistent with the results from an independent model intercomparison (AeroCom; Aerosol Comparison between Observations and Models). The new scheme is also found to produce black carbon surface concentrations resulting from long range transport that are in better agreement with other models than the standard scheme.

Introduction

Black carbon abundance is important for both the climate and for air quality. In the climate system, black carbon interacts directly with solar radiation both through absorption and scattering. It also may have indirect effects on cloud albedo, abundance and lifetime (e.g. Haywood and Boucher, 2000). When transported to the arctic regions and deposited on snow surfaces, black carbon can change the snow albedo (e.g. Flanner et al., 2007). Thus, black carbon has a complex multifaceted effect on climate. Black carbon additionally contributes to surface particulate matter concentrations which are typically quantified as PM2.5 and PM10 levels. PM2.5 and PM10 has a direct impact on human health due to inhalation (e.g. WHO, 2006).

The abundance of black carbon in the atmosphere is determined by three types of process: the rate of emission from sources, transport in the atmosphere and removal from the atmosphere due to wet and dry processes. A useful concept is the atmospheric residence lifetime as it relates directly to the burden. When an aerosol for a given set of conditions reaches a steady state, the lifetime can be calculated as follows:

$$Lifetime = \frac{Burden}{Emission\ rate} \quad ,$$

where the burden is the total mass of the aerosol in the atmosphere (often expressed in Tg) and the emission rate (for primary aerosols) is the average rate of input of the substance into the atmosphere over a given time period (e.g. Tg year^{-1}) . For aerosols, it is convenient for to express the resultant lifetime in units of days.

CLASSIC is a mass based aerosol module commonly used in the Hadley Centre Global Environmental Model, currently implemented in versions HadGEM2 and HadGEM3 (Bellouin et al., 2011). It is known that black carbon in the standard CLASSIC scheme has a long lifetime when compared to other studies (Bellouin et al, 2012 and Koch et al.,

2009). A long lifetime will typically enhance long range transport leading to enhanced aerosol optical depths and concentrations in remote regions.
The aerosol lifetime is dependent on the strength of the removal processes; particularly by wet deposition. Wet deposition in CLASSIC occurs during precipitation of droplets containing aerosol mass. Black carbon in CLASSIC is treated as a hydrophobic aerosol. Thus, the only way for the aerosol to enter a droplet is through diffusional scavenging; an aerosol particle has to physically impact a droplet to enter it. This is a much slower process than for hydrophilic aerosols such as sulphate where nucleation scavenging can occur and a particle can form a droplet of its own. Thus it follows that by increasing the black carbon hygroscopicity and making it able to participate in nucleation scavenging, the lifetime will reduce. This is physically plausible due to aging of the emitted black carbon. Although the BC is treated as an external mixture in CLASSIC, in reality it will combine with other more hydrophilic particles over time.

Experimental method

Increasing the hygroscopicity of black carbon is implemented in a new scheme by replacing the removal mechanism with that used for the other carbonaceous aerosols in CLASSIC (namely organic carbon from fossil fuel burning and biomass burning). These aerosols are somewhat hydrophilic, but less so than sulphate (See appendix of Bellouin et al., 2011). It is important to note that in this new method, the cloud scheme does not consider black carbon as cloud condensation nuclei, even when coupling between aerosols and climate is enabled. This is to minimise the differences in aerosol indirect effects compared to the standard scheme. Parallel experiments are conducted to compare the the new black carbon removal scheme with the standard scheme. One pair of experiments has climate and emissions appropriate for the present day. A further two pairs only consider black carbon emissions from North America and Europe and investigate the effect of long range transport on black carbon surface concentration. The experiments use seasonally evolving annually repeating emissions and sea surface temperatures for a decadal average centered on the year 2000 from the HadGEM2 CMIP5 runs (Bellouin et al., 2011). All experiments each simulate 5 years after 13 months of spin up. Interaction between the aerosols and the climate (via direct and indirect effects) is inhibited in order to maintain consistent meteorology between the experiments. For surface concentrations, an average of the the first 5 model levels (from the surface to ~500m) is used to allow for vertical mixing in the boundary layer.

The source regions are defined as global, North America (15N to 55N; 125W to 60W) and Europe (25N to 65N; 10W to 50E); using the HTAP (2010) specification as a basis. Europe is also used as a receptor region in addition to a separate smaller receptor region over the UK (49.75N to 60N; 10W to 2.5E). Note that these are simple broadly defined regions and do not represent geopolitical boundaries. The “European region” contains parts of North Africa for example.

Results

The atmospheric lifetime for black carbon is determined for CLASSIC using both the standard removal and new removal schemes. These lifetimes are compared to those from an AeroCom aerosol model intercomparison that consisted of 12 model members (Table 1). The black carbon in standard scheme has a long lifetime of 16.2 days globally averaged. This lies well outside of the range of the AeroCom ensemble (upper value of 11.4 days). When the new black carbon scheme is used, the lifetime becomes 6.0 days for global emission. This is very close to the AeroCom median of 6.4 days and within 1 standard deviation of the mean value of 6.7 days. There is also greater dependence on the source region with the new scheme. Emissions from North America and Europe lead to shorter lifetimes than the global mean. This is due to the inhomogeneous nature of global cloud and precipitation patterns in relation to black carbon source regions, removal in these in areas being more efficient than in the global mean. Figure 1 shows the resultant distribution of black carbon burden in the Northern Hemisphere. When using the new scheme, there is a large effect on BC burden remote from the source regions when compared to the standard scheme. For example, there is an order of magnitude less aerosol reaching the the Arctic from European sources.

	Emission source region		
Experiment	Global	North America	Europe
AeroCom Models (Koch et al., 2009)	Mean : 6.7 Median: 6.4 Min : 4.9 Max : 11.4 *Sigma : 1.7*	---	---
CLASSIC with standard BC removal	16.2	16.0	16.0
CLASSIC with new BC removal	6.0	4.8	5.5

Table 1: Comparison of black carbon lifetimes (in days).

Different lifetimes of black carbon result in different surface concentrations, especially at locations remote from the source (Figure 2). To investigate this, the contribution of North American black carbon emissions to UK and European average surface concentration levels are determined. The contributions determined from the standard and new schemes are compared with a Hemispheric Transport of Air Pollution (HTAP, 2010) multi-model ensemble (Table 2). By using the new scheme, CLASSIC has a European surface concentration contribution from North America that is within the range of the HTAP ensemble. The standard scheme gives a substantially larger surface concentration contribution, falling outside of the upper bound of the HTAP ensemble and ~4x greater than the median.

When compared to the European region, the UK experiences a greater surface concentration contribution from North America in the CLASSIC experiments. This is due to the exact positioning of the North American plume, with the plume center passing close to the UK (Figure 2) and the proximity of the UK to North America relative to the European region mean.

Experiment	UK average	European average
HTAP Models (HTAP, 2010)	---	Median: 0.0029 Min : 0.0003 Max : 0.0103
CLASSIC with standard BC removal	0.0127	0.0110
CLASSIC with new BC removal	0.0013	0.0011

Table 2: Comparison of BC surface concentration contributions (in $\mu g\ m^{-3}$) resulting from North American emissions. The range in HTAP values is calculated from the range in absolute surface concentrations and the range in percentage contributions from North America (see Table 4.6 of HTAP, 2010).

Conclusions and recommendations

It has been shown that increasing the hygroscopicity of black carbon in CLASSIC results in a shorter atmospheric lifetime of ~6 days globally that is consistent with other studies. This in turn reduces the long range transport of black carbon and results in a lower fraction of black carbon aerosol found in surface concentrations of receptor regions.

The primary use of the new scheme is for air quality studies to avoid overestimation of surface level particulate matter, particularly from remote sources. Although not a focus of this study, the new scheme should also be considered for studies involving black carbon deposition as often important receptor regions are remote from the source. For example, the sensitivity of the Arctic to transport of black carbon from Europe (e.g. Lee et al., 2012).

Caution is required if using the new scheme when aerosols are coupled to the climate. Although aerosol indirect effects are unchanged relative to the standard scheme, the change in black carbon burden will introduce differences in the global distribution of direct and semi-direct effects. Additionally, if the impacts of BC on snow albedo are to be included, there would be large differences in forcing over the Arctic regions between the two schemes.

The new black carbon removal scheme is available for both HadGEM2 and HadGEM3 when using the CLASSIC aerosol module. The scheme is not compatible (or indeed required) when using the GLOMAP-mode aerosol module which already has a black carbon lifetime comparable with other studies (Bellouin et al., 2012).

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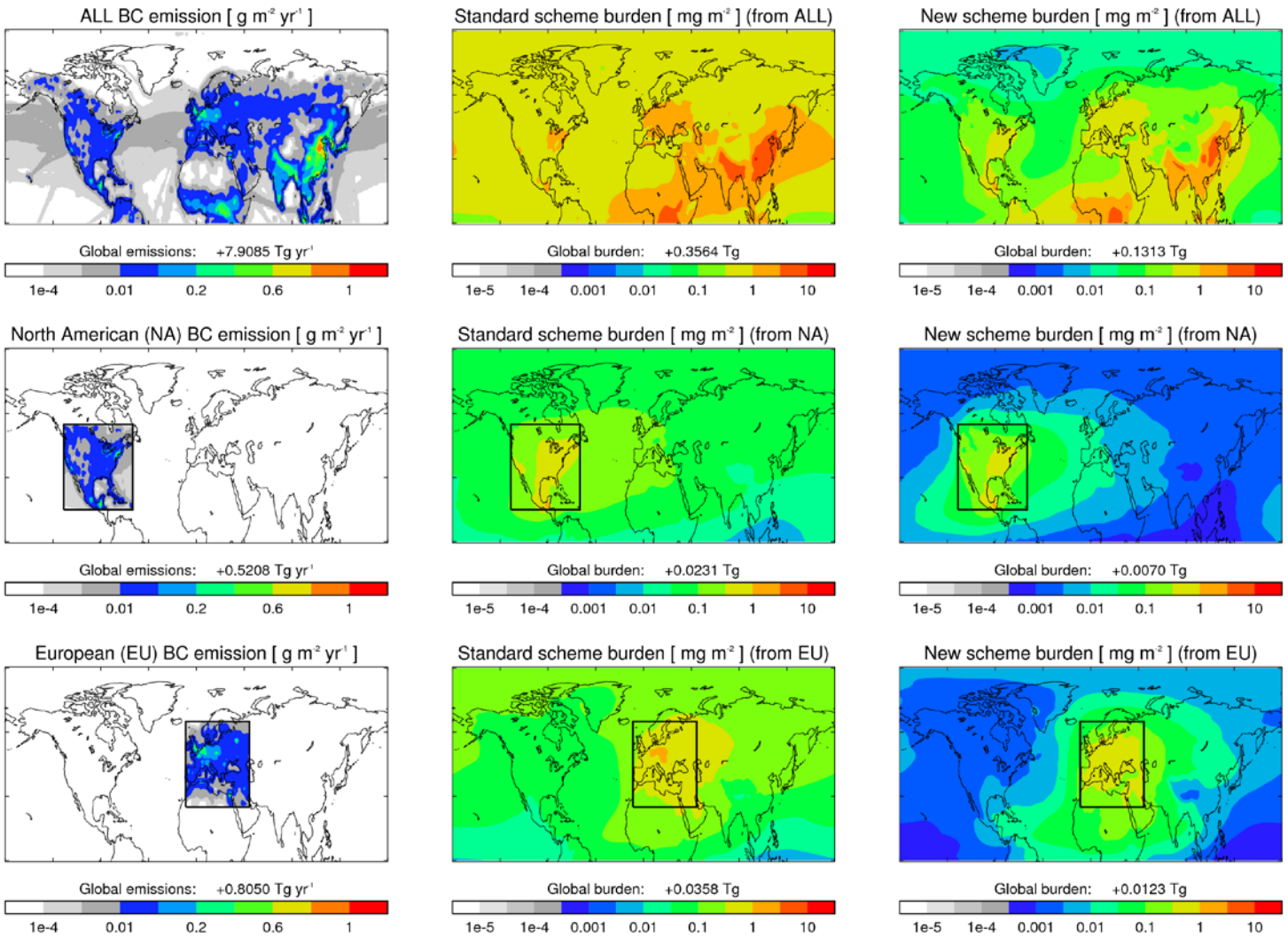

Figure 1: Black carbon burden comparison between schemes (standard scheme, centre; new scheme, right). Left hand column are the emissions for reference. Top row are from global emissions. Middle row are from North American emissions and bottom row are from European emissions. Global values are for both hemispheres. Graphics depict Northern Hemisphere only. See text for NA, EU and UK region definitions. All are log scale apart from emissions 0.2 g m^{-2} yr^{-1} and greater which are linear.

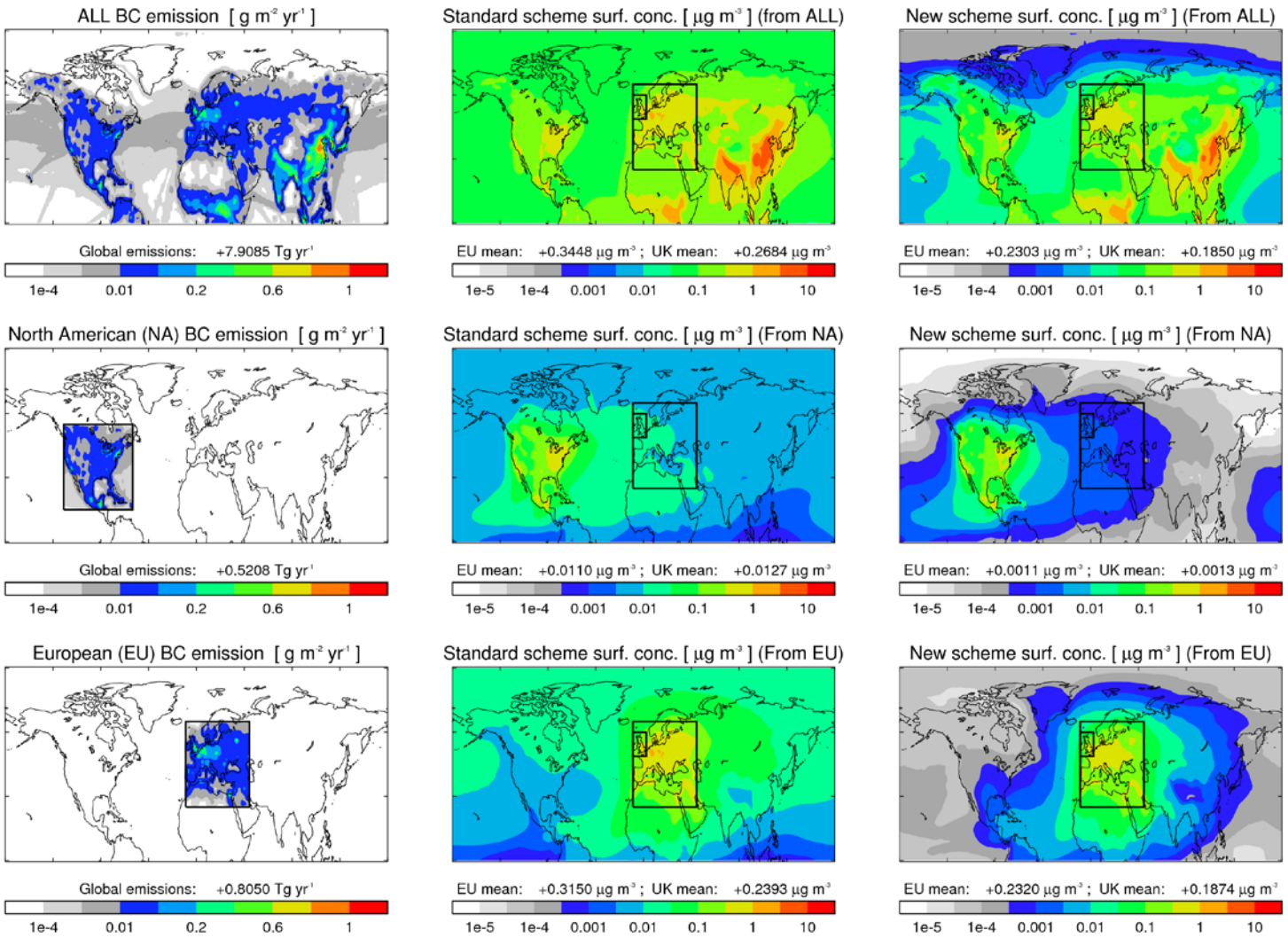

Figure 2: As in Figure 1, but with surface concentrations instead of burdens. EU and UK receptor regions bound by boxes