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Refractory black carbon at the Whistler Peak High Elevation Research Site – Measurements and simulations

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ABSTRACT

Measurements of black carbon at remote and high altitude locations provide an important constraint for models. Here we present six months of refractory black carbon (rBC) data collected in July–August of 2009, June–July of 2010, and April–May of 2012 using a single particle soot photometer (SP2) at the remote Whistler High Elevation Research Site in the Coast Mountains of British Columbia (50.06°N, 122.96°W, 2182 m a.m.s.l). In order to reduce regional boundary layer influences, only measurements collected during the night (2000-0800 PST) were considered. Times impacted by local biomass burning were removed from the data set, as were periods of in-cloud sampling. Back trajectories and back trajectory cluster analysis were used to classify the sampled air masses as Southern Pacific, Northern Pacific, Western Pacific/Asian, or Northern Canadian in origin. The largest rBC mass median diameter (182 nm) was seen for air masses in the Southern Pacific cluster, and the smallest (156 nm) was seen for air masses in the Western Pacific/Asian cluster. Considering all the clusters, the median mass concentration of rBC was 25.0 ± 7.6 ng/m^3-STP. The Northern Pacific, Southern Pacific, Western Pacific/Asian, and Northern Canada clusters had median mass concentrations of 25.0 ± 7.6, 21.3 ± 6.9, 25.0 ± 7.9, and 40.6 ± 12.9 ng/m^3-STP, respectively. We compared these measurements with simulations from the global chemical transport model GEOS-Chem. The default GEOS-Chem simulations overestimated the median rBC mass concentrations for the different clusters by a factor of 1.2–2.2. The largest difference was observed for the Northern Pacific cluster (factor of 2.2) and the smallest difference was observed for the Northern Canada cluster (factor of 1.2). A sensitivity simulation that excluded Vancouver emissions still overestimated the median rBC mass concentrations for the different clusters by a factor of 1.1–2.0. After implementation of a revised wet scavenging scheme, the simulations overestimated the median rBC mass concentrations for the different clusters by a factor of 1.0–2.0.

1. Introduction

A major component of the particulate matter formed during fossil-fuel, biomass, and biofuel combustion is the light-absorbing, refractory, and insoluble material known as black carbon (BC) (Bond et al., 2013; Petzold et al., 2013). The impact of black carbon in the atmosphere is complex. It absorbs both incoming and outgoing radiation, which can lead to a reduction in the amount of light reaching the surface and to a warming of the atmosphere. This has an impact in terms of direct radiative forcing and also has the potential to significantly perturb the hydrological cycle (Bond et al., 2013; Ramanathan and Carmichael, 2008; Samset et al., 2013). Particles containing black carbon may also act as cloud condensation nuclei (Kotzick et al., 1997; Petzold et al., 2005; Schroder et al., 2015; Zuberi et al., 2005), which can lead to

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Fig. 1. Location of the Whistler Peak High Elevation Research Site (red star). The locations of Vancouver and Seattle (blue triangles), and the GEOS-Chem grid box used (grey area) are also shown. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

changes in the radiative properties of clouds (Chen et al., 2010; Koch et al., 2011). In addition, black carbon impacts climate through deposition on snow or ice surfaces, reducing their albedo and increasing their absorption of solar radiation (Clarke and Noone, 2007; Flanner et al., 2009; Hadley and Kirchstetter, 2012; Ramanathan and Carmichael, 2008).

It is important that black carbon is accurately represented in models because of its importance in climate and in the hydrological cycle. As a result, measurements of BC (for example Dahlkötter et al., 2014; Kondo et al., 2011; Liu et al., 2013; McMeeking et al., 2010; Metcalf et al., 2012; Reddington et al., 2013; Schwarz et al., 2010a; Wang et al., 2014a) and comparison of measurements with model predictions have been a focus of recent studies (Kipling et al., 2013; Koch et al., 2009; Reddington et al., 2013; Schwarz et al., 2010a, 2013; Wang et al., 2014b; Zhang et al., 2015). The comparisons between measurements and models have revealed that models often overestimate BC in remote regions and the free troposphere. For example, Koch and colleagues performed a detailed comparison of available BC measurements with predictions from the AeroCom (Aerosol Comparisons between Observations and Models) suite of global models, and they found that the simulated total column burden (excluding lowest 2 model layers) was on average a factor of 8 larger than aircraft measurements over the Americas at latitudes between 0 and 50N (Koch et al., 2009). Schwarz and colleagues compared black carbon measurements over the remote Pacific from the HIAPER Pole-to-Pole Observations (HIPPO) campaign in January 2009 with results from the AeroCom suite of models, and they found that the median model concentration was greater than the observed BC concentration by a factor of five on average (Schwarz et al., 2010a). Kipling et al. (2013) compared measurements of BC over the remote Pacific from HIPPO during 2009–2010 with two global models, HadGEM3–UKCA and ECHAM5–HAM2. They showed that the models often overestimated BC measurements in remote regions, in some cases by an order of magnitude (Kipling et al., 2013). A different study comparing BC measurements from the 2009–2011 HIPPO campaign over the remote Pacific with a GEOS-Chem simulation showed that the simulation had a high bias relative to the measurements of approximately a factor of two (Wang et al., 2014b).

Additional measurement-modelling comparisons of BC are needed to test the skill of models for predicting BC. Model development informed by the measurement-modelling comparisons should eventually lead to better representations of processes affecting BC in simulations. Here we present six months of refractory black carbon measurements in periods spanning 3 years at the Whistler Peak High Elevation Research Site in the Coast Mountains of British Columbia (50.06°N, 122.96°W, 2182 m a.m.s.l). Whistler Peak is a remote station, which is often in the lower free troposphere (D'Andrea et al., 2016) and sees little influence from any large urban centers (Macdonald et al., 2011). In addition, transpacific transport from Asia has been observed to impact this site (Mckendry et al., 2007; Sun et al., 2009), and plumes are also transported above the site (Leaitch et al., 2009).

The first goal of this study is to measure mass concentrations and size distributions of refractory black carbon at this remote, high altitude site. Since burdens of black carbon in remote and high altitude locations can be especially sensitive to sinks of BC and can be sourced from multiple regions, measurements at remote and high altitude locations provide an important constraint for models. In addition, these measurements should provide useful information on background levels and microphysical properties of black carbon in the Pacific Northwest region which is important when trying to distinguish between regional and background sources. The second goal of this study is to assess the level of skill of the chemical transport model GEOS-Chem at predicting concentrations of black carbon at this remote and high altitude location. In addition, we test the wet scavenging developments of Wang et al. (2014b) in this chemical transport model to examine their effect on agreement between measured and simulated black carbon mass concentrations at Whistler Mountain.