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Formation and evolution of tar balls from northwestern US wildfires

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Supplemental Information

Thermal Stability of Tar Balls and their Detection by an SP-AMS

The stability of TBs during particle collection and subsequent electron microscopy suggests that they exhibit a significant physical and thermal robustness that may impact their detection by instruments that exploit particle volatility (including SP-AMS, measurements obtained from thermal denuders, and by thermal/optical carbon analyzers. See Adachi et al., 2017). Two experiments were conducted in order to evaluate the thermal robustness of TBs and a third to determine the CE of TBs by an SP-AMS. The first was a proof-of-principal study in which a TEM grid (sampled at between 19:04:48 and 19:13:12, July 19 (UTC) from the Colockum Tarp wildfire was subjected to a 15-minute heat ramp from room temperature to 650 ºC. TEM images (Figure S1) collected at 30 ºC, 200 ºC, 400 ºC, and 600 ºC show that TBs and ns-soot remain, although ~70% of the volume of the TBs was lost by 600 ºC (Figure S2). In the second experiment described by Adachi et al. (2017), a slightly revised protocol (10-minute heat ramp to 600 ºC) was used to determine TB shrinkage as a function of temperature using nine TEM grids from two fire plumes and an agricultural burn in Arkansas, all sampled during the BBOP field campaign. It is to be noted that TBs typically accounted for < 1% of particle composition distribution in agricultural burns studied with TEM. TB volume loss in the wildfire samples was nearly identical to that shown in Fig. S2. The heating profiles in these experiments were very different than the flash vaporization that occurs when aerosol hits the 600 ºC oven within an SP-AMS.

A third set of laboratory experiments to specifically address the response of an SP-AMS to TBs was conducted (Onasch et al., 2017). Detection efficiency of TBs could be limited by the low volatility of TBs and by particle bounce of semi-solid and/or highly viscous particles impacting the heated tungsten vaporizer (Cross et al., 2009; Matthew et al., 2008). Procedures outlined in Tóth et al. (2014) and Hoffer et al. (2016) were used to generate laboratory TBs. Preliminary results indicate that the SP-AMS used during the BBOP study can detect laboratory-generated TBs with a CE similar to that observed for most ambient conditions (Hoffer et al., 2016). These limited laboratory experiments on laboratory-generated TBs provide some confidence in the SP-AMS OA and TB measurements during BBOP.

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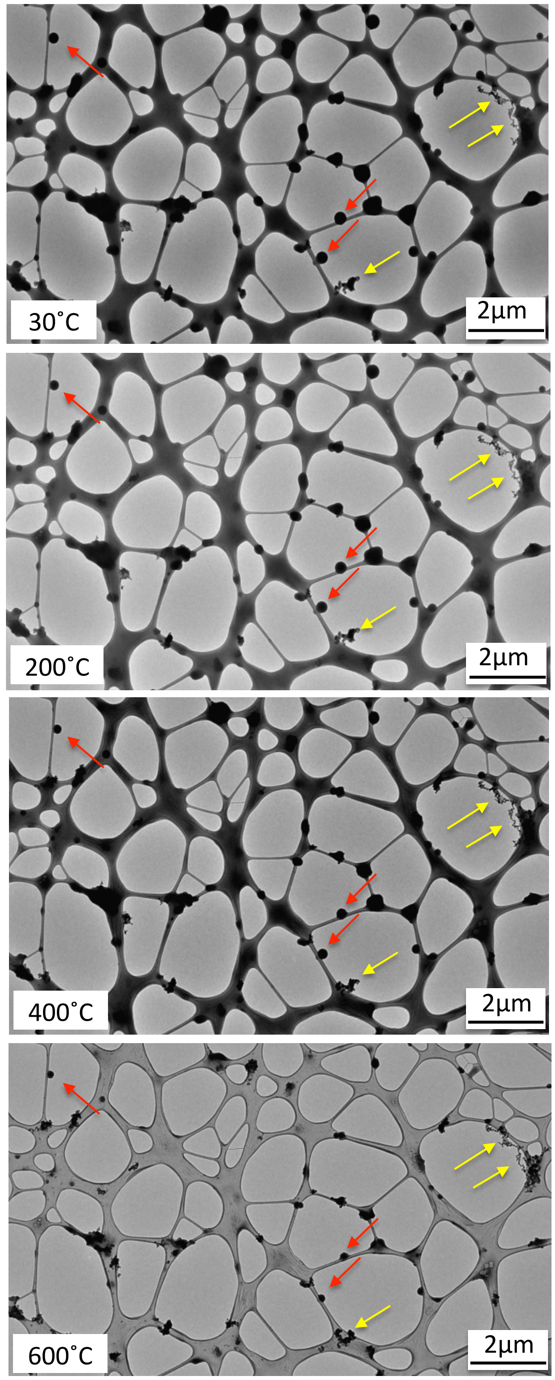

Figure S1. Thermal robustness of TBs. A BBOP TEM grid was heated in 50 °C increments over the course of 15 minutes to evaluate the thermal stability of TBs (red arrows). Although some loss of material occurs, it is clear that TBs exhibit significant thermal stability. Yellow arrows indicate ns-soot.

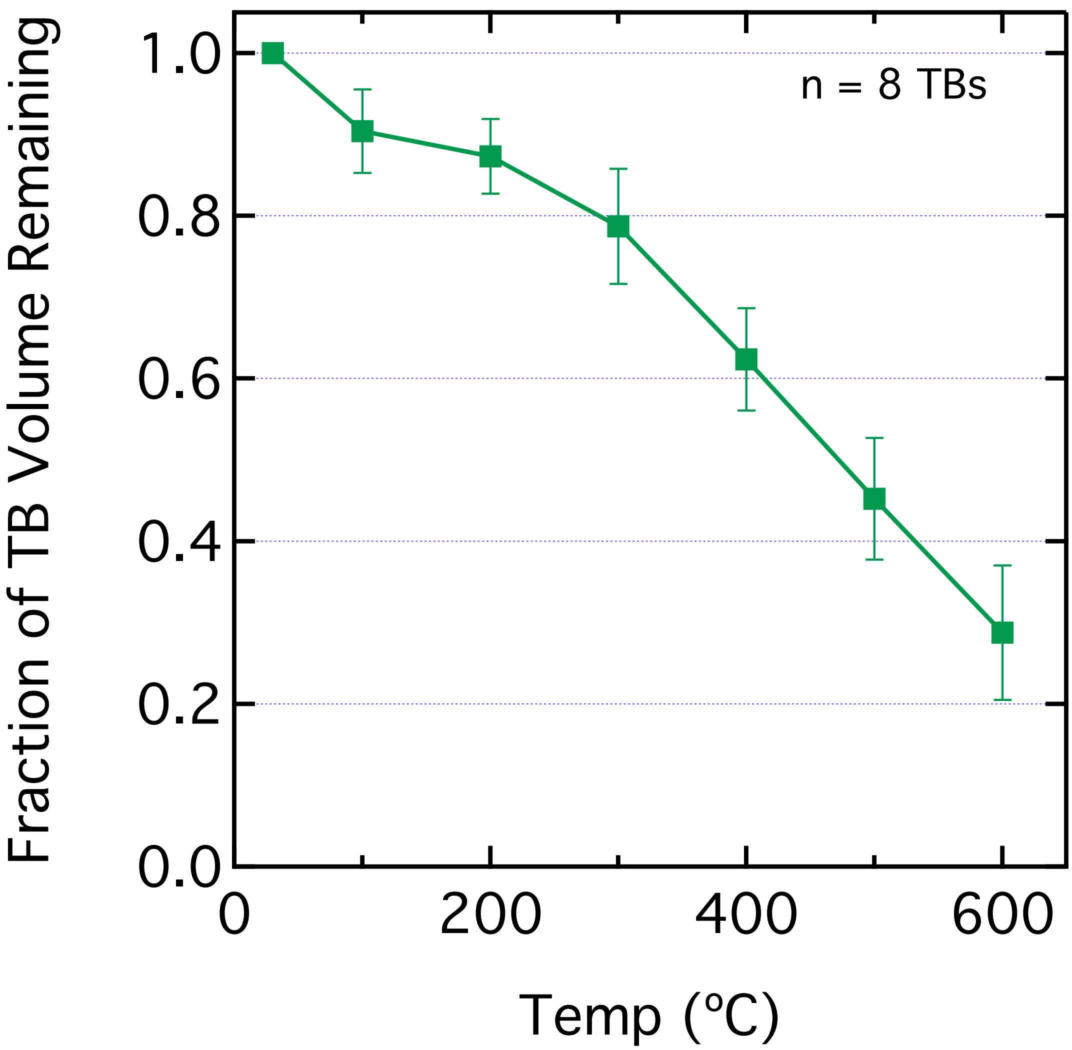

Figure S2. Volume fraction of TBs remaining upon heating in the electron microscope. Temperature is the calculated temperature in the furnace within TEM. These particles are shown in Figure S1. The volumes were estimated from the projected areas in the TEM images, assuming spherical shapes.