

# Consistency of Long-term Black Carbon Trends from Thermal and Optical Measurements in the U.S. IMPROVE Network

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Decreasing trends of black carbon (BC), also termed elemental carbon (EC) as determined by thermal/optical analysis, have been reported at the U.S. IMPROVE (Interagency Monitoring of PROtected Visual Environments) network from 1990 to 2004 (Murphy *et al.*, 2011), consistent with the phase-in of cleaner engines, residential biomass burning technologies, and prescribed burning practices. BC trends for the past decade (2000–2009) are examined in this study due to an upgrade of IMPROVE carbon instruments and the thermal/optical analysis protocol since 2005.

The IMPROVE thermal/optical reflectance (TOR) method separates BC from organic carbon (OC) on filter samples by temperature-dependent volatilization and oxidation. A fraction of OC chars in the inert atmosphere, as evidenced by decreases in light (632.8 nm He/Ne laser) reflected from the aerosol deposit on the filter surface, is continuously monitored during the analysis and corrected from BC measurements (Chen *et al.*, 2004). The 2005 carbon instrument upgrade led to a transition from the IMPROVE to IMPROVE\_A protocol (Chow *et al.*, 2007). The new protocol does not change the temperatures plateaus but rather reflected the “actual” analysis temperatures that had been implemented since the inception of the IMPROVE network. It also enables simultaneous monitoring of filter reflectance and transmittance without changing the reflectance measurement configuration.

Optical measurements designed for charring correction provide alternatives for quantifying BC abundances on filters. Filter attenuation using reflected light ( $\tau_R$ ) is defined as

$$\tau_R = -\ln(R/R_0) \quad (1)$$

where  $R$  and  $R_0$  are the reflected light intensity of particle-laden (prior to carbon analysis) and blank (after carbon analysis) filter, respectively.  $\tau_R$  is an independent measure of light absorption coefficient ( $b_{abs}$ ) as well as BC on the filter (e.g., Janssen *et al.*, 2011). Digital thermograms of > 83 000 IMPROVE samples acquired by 24-h sampling on every third day from 2000 through 2009 were reprocessed to obtain the  $\tau_R$  values. The 65 sites with the longest records and highest data recovery rates are used for this analysis (Chen *et al.*, 2012). They represented 25 U.S. geographic regions.

The changes in regression coefficients between BC and  $\tau_R$  before and after 2005 were first examined with a dummy-variable approach (Gujarati, 1970), and relationships between BC measured from the two periods were established (Chen *et al.*, 2012). The variability is shown to be within  $\pm 10\%$  in general. Larger deviations occur for high or low extreme BC levels.

BC and  $\tau_R$  trends were further assessed using a nonparametric Mann-Kendall (M-K) test. M-K statistics yield Sen's slope (Burn and Hag Elnur, 2002) and its p-value and confidence intervals. BC and  $\tau_R$  show universal decreasing trends across the U.S. The BC and  $\tau_R$  trends are well correlated (Figure 1), with national average downward rates (relative to the 2000–2004 baseline medians) of 4.5 % yr<sup>-1</sup> for BC and 4.1 % yr<sup>-1</sup> for  $\tau_R$ . The consistency between BC and  $\tau_R$  measurements adds to the weight of evidence that BC reductions are real rather than an artifact of changes to the measurement process. At the current rate of progress, the goal of the U.S. Regional Haze Rule for BC should be met by the 2064 deadline.

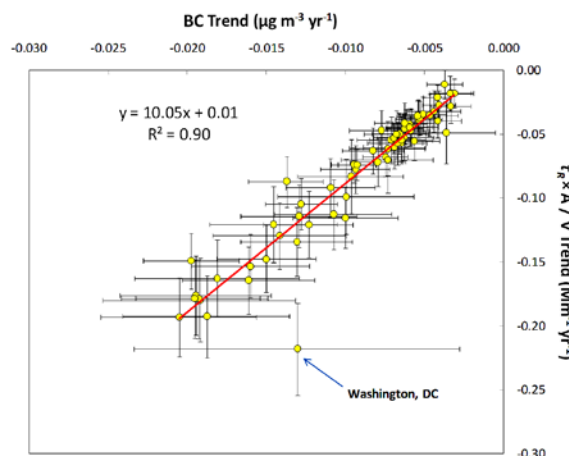


Fig. 1. A comparison of BC and  $\tau_R$  trends for 65 IMPROVE sites during 2000–2009. The unit of slope is  $\text{m}^2 \text{g}^{-1}$ , corresponding to mass specific absorption of BC.

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