Table S1. Sign table used in the HSRI-2 retrieval algorithm to disturb the input data (i.e. $3\beta + 2\alpha$) with their respective uncertainties. For instance, the first run of the inversion algorithm starts with an input set comprised of $\alpha_{355} + \Delta \alpha_{355}$, $\alpha_{532} + \Delta \alpha_{532}$, $\beta_{355} + \Delta \beta_{355}$, $\beta_{532} + \Delta \beta_{532}$, $\beta_{1064} - \Delta \beta_{1064}$, where Δs refer to uncertainties.

Run number	α_{355}	α_{532}	β_{355}	β_{532}	β_{1064}
1	+	+	+	+	-
2	-	-	+	+	-
3	+	-	+	+	-
4	-	+	+	+	-
5	+	+	-	-	+
6	-	-	-	-	+
7	+	-	-	-	+
8	-	+	-	-	+



Figure S1. Comparison of AOT measurements at 355 and 532 nm measured with HSRL-2 and AERONET/DRAGON sunphotometers during DAQ California (left) and DAQ Texas (right). Measurements from AERONET were calculated from level 2.0 measurements of AOT at 340 nm and 500 nm, corrected to 355 and 532 nm using Ångström exponents calculated between 340 and 440 nm, and 440 and 675 nm, respectively. Only measurements obtained within a radius of 2.5 km and 10 minutes from each other were considered.



Figure S2. In situ AOT fraction calculated from AERONET and in situ measurements vs. AERONET Ångström exponent calculated from AOT measurements at 440 nm and 870 nm.



Figure S3. Median extinction profiles measured with HSRL-2 at 532 nm during DAQ in California (Blue) and Texas (Red). Shaded areas represent the 25th and 75th percentiles.