

An observationally constrained framework for black-carbon microphysics, column loading and radiative forcing: from regional hotspots to global applications

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Black carbon (BC) remains one of the most uncertain climate forcers because its direct radiative effect depends non-linearly on per-particle microphysical state (size, mixing state, core-shell morphology) and on column loading, not only on mass. Traditional retrievals that assume fixed size distributions or Angstrom-exponent-based source attribution cannot capture this complexity. This leads to physically unrealistic (negative or implausibly small or large) emission estimates.

We have built a multi-platform, multi-waveband, observationally constrained inversion framework (COSMO) that explicitly accounts for BC microphysical variability and propagates observational uncertainty through the full inversion chain. This newest version treats the aerosol population as an external mixture of three physically distinct archetypes: (1) BC-only, (2) BC + brown carbon (BrC), (3) BC + ionic material (coal-ash type). Core-shell Mie calculations span realistic ranges of core radius (10–250 nm) and shell thickness (5–990 nm). Effective refractive indices of mixed cores are computed using Maxwell-Garnett.

Observational constraints are applied sequentially:

- Spectral single-scattering albedo (SSA) from AERONET/SONET, TROPOMI, OMI, & MISR filters particles with inconsistent scattering behaviour.
- AAOD determines column number densities of each archetype.
- Total AOD (TROPOMI, MODIS MAIAC) and degree of linear polarization from GF-5 DPC reject contaminated solutions (coarse dust or ice), retaining only combustion-derived aerosols.

The final ensemble provides day-by-day, grid-by-grid column mass, number densities, and microphysical properties per particle, including explicit uncertainty quantification. This in turn is used to drive SBDART radiative transfer calculations for top-of-atmosphere and atmospheric forcing and radiance.

In urban/industrial hotspots (Xuzhou, Dhaka, Shanxi coal basins), we show that particle size distribution, not only mass, controls the spectral behaviour of SSA and asymmetry parameter. Ignoring this introduces systematic bias in both AOD and AAOD, and can change estimated radiative forcing by a factor of two, or even reverse its sign.

We demonstrate thick coatings simultaneously increase BC absorption (lensing effect) and scattering (due to the shell's refractive index). This dual effect leads to a non-linear, sign-variable change in top-of-atmosphere radiance across the solar spectrum, with impacts also in the short-wave infrared (2.3–2.4 μm), a region previously assumed by the methane community to be free of aerosol interference.

Using MISR multi-band observations (443–865 nm) we have produced the first global, daily, 0.5° resolution BC dataset, including column mass, number, effective radius and mixing-state information. The dataset reveals strong spatial heterogeneity and contrasting trends: in western

China BC mass has decreased due to policy actions, but particle size and mixing state have not followed simple trends.

The same coating process also biases satellite methane retrievals: aerosol-induced radiance perturbations translate into equivalent XCH_4 biases of up to ± 50 ppb, comparable to or larger than TROPOMI's stated precision. In coal-dominated regions like Shanxi, methane emission estimates can be biased by 20% or more, affecting greenhouse gas inventories.

Relevance to GRASP

Our work shares strong methodological synergies with GRASP: multi-platform, multi-angle, multi-spectral inversion with explicit uncertainty propagation. We would like to couple COSMO-derived a priori particle size and mixing state distributions into a GRASP-like retrieval chain. We also hope that our derived BC microphysical datasets can serve as an independent constraint for aerosol typing and model evaluation.