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Particle-Resolved Compositional Fingerprinting of Brake Wear Particles by spICP-TOFMS: Implications for Urban Source Attribution

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Non-exhaust emissions from vehicles, particularly brake wear particles, are increasingly recognized as a dominant source of urban particulate pollution, yet accurate source attribution remains challenging. Conventional bulk analysis averages across compositionally distinct particle populations, obscuring the element associations that carry source information at the individual particle level. Single-particle inductively coupled plasma time-of-flight mass spectrometry (spICP-TOFMS) offers a powerful alternative, enabling quasi-simultaneous multi-element characterization of individual particles at high throughput.

We analyzed brake wear particles collected from 49 in-use tire rims across Vienna, Austria. Unlike laboratory-generated brake material, these samples represent brake emissions accumulated during real-world vehicle operation, providing a more realistic basis for source characterization. Sample preparation followed optimized protocols including FL-70 surfactant stabilization and vial-tweeter sonication. The spICP-TOFMS configuration enabled detection of 21 elements across close to 1.6 million individual particles, a scale not feasible with conventional approaches.

Analysis of the resulting dataset revealed that traditional bulk brake markers (Fe, Cu, Ba, Sb) rarely co-occur at the single-particle level, despite their widespread use in bulk source attribution. Instead, the dataset is dominated by pure Fe particles, with Fe-Sn associations representing the second most common pattern. Unsupervised clustering of particle compositions revealed two broad groupings: brake-related populations characterized by Fe with varying contributions of Cu, Sn, Cr, and other brake tracers, and environmentally-influenced populations distinguished by enrichment in Si and Al.

These results demonstrate that particle-resolved fingerprinting by spICP-TOFMS, combined with unsupervised classification, improves source attribution of brake wear particles. By resolving compositional complexity that bulk approaches cannot capture, this framework provides a more informative basis for tracking the environmental fate and health impacts of non-exhaust emissions in urban environments.