Supporting Information

Novel AI Technology for 4D In-situ Tracking and Physicochemical Characterization of Inorganic Carbonaceous Aerosols in Air/Water

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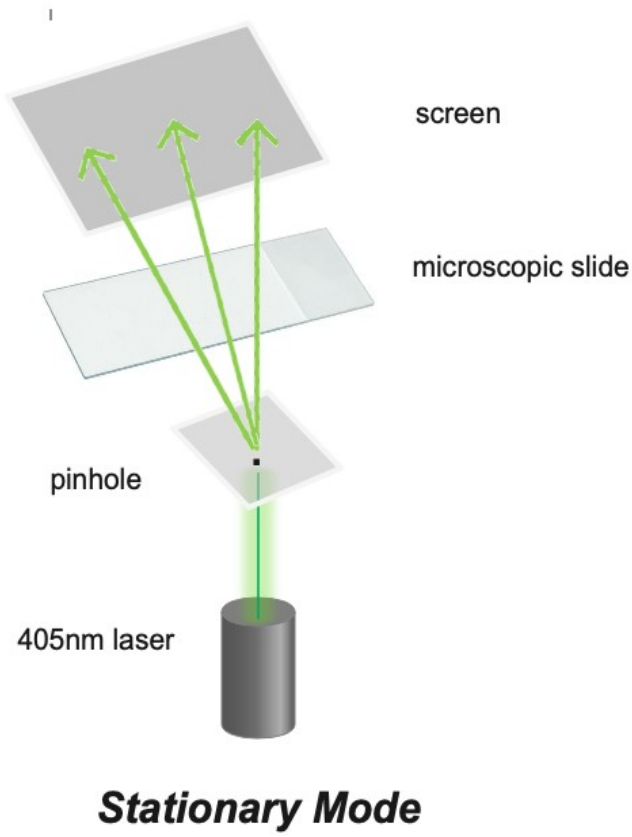
Supplementary Figures

a)



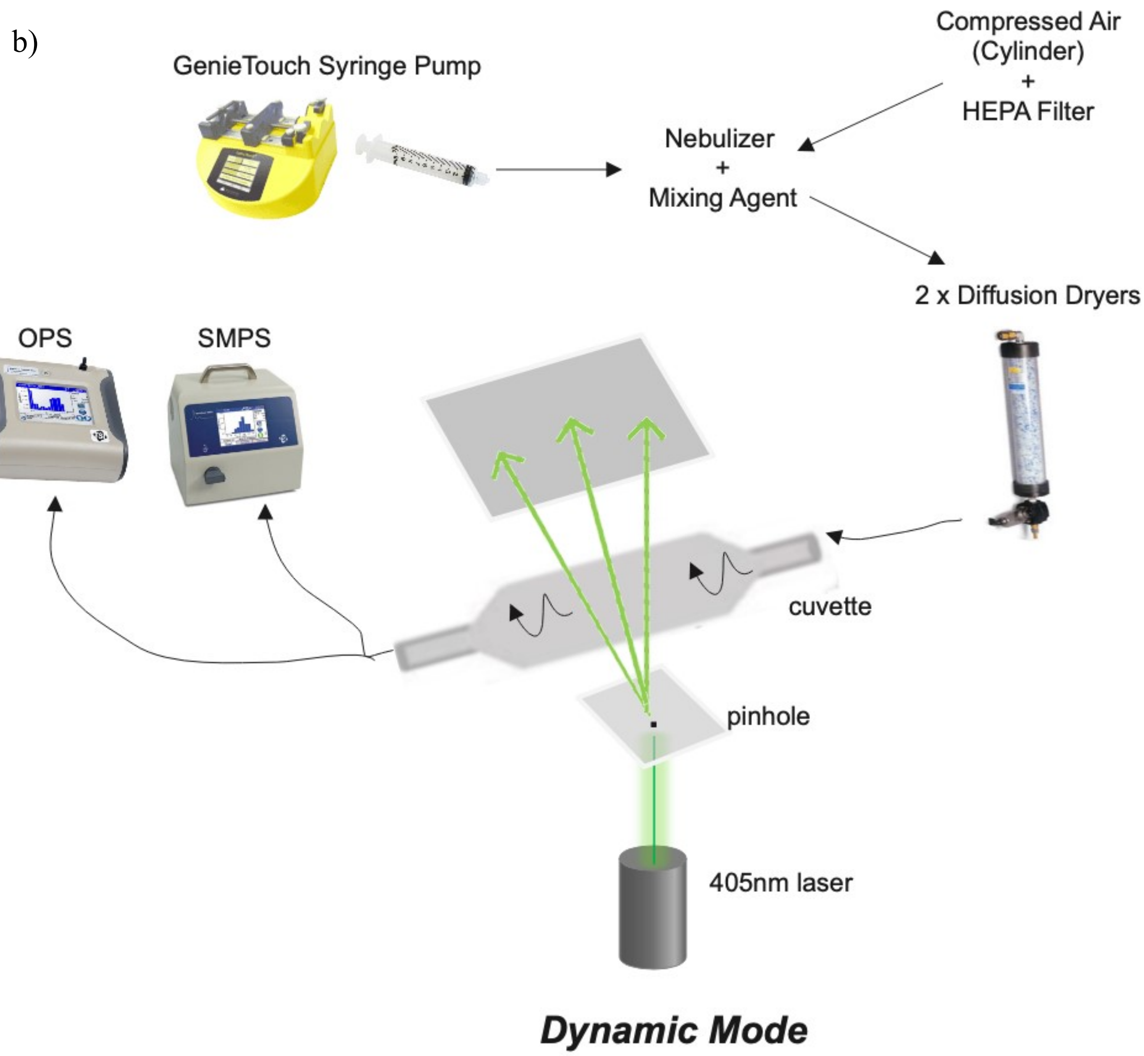


Figure S1. (a) Stationary mode setup, (b) Dynamic mode setup

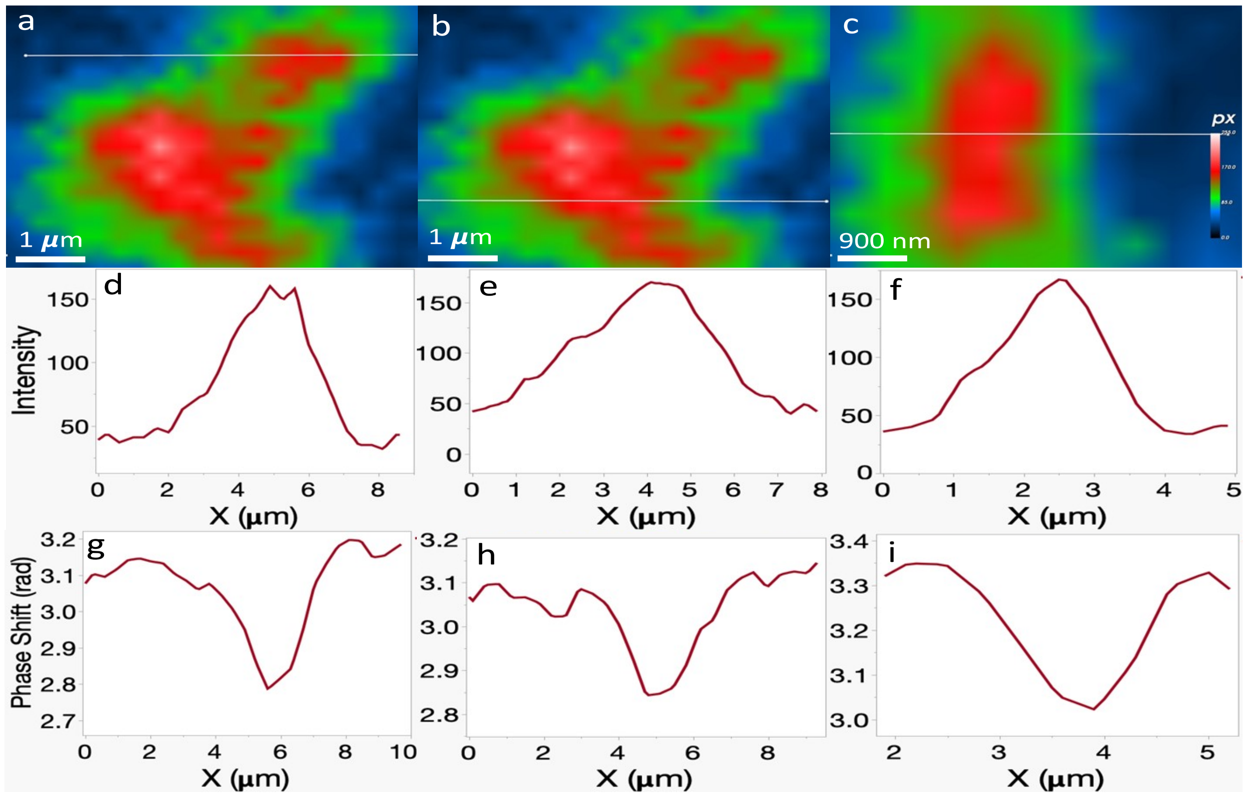


Figure S2. Reconstruction of the graphite particles in the water phase. (a-c) Intensity reconstruction of graphite particles, (d-f) Intensity profiles of particles 1 and 2 across the particles crosscut, (g-i) The phase profiles of particles 1 and 2 across the particles crosscuts.

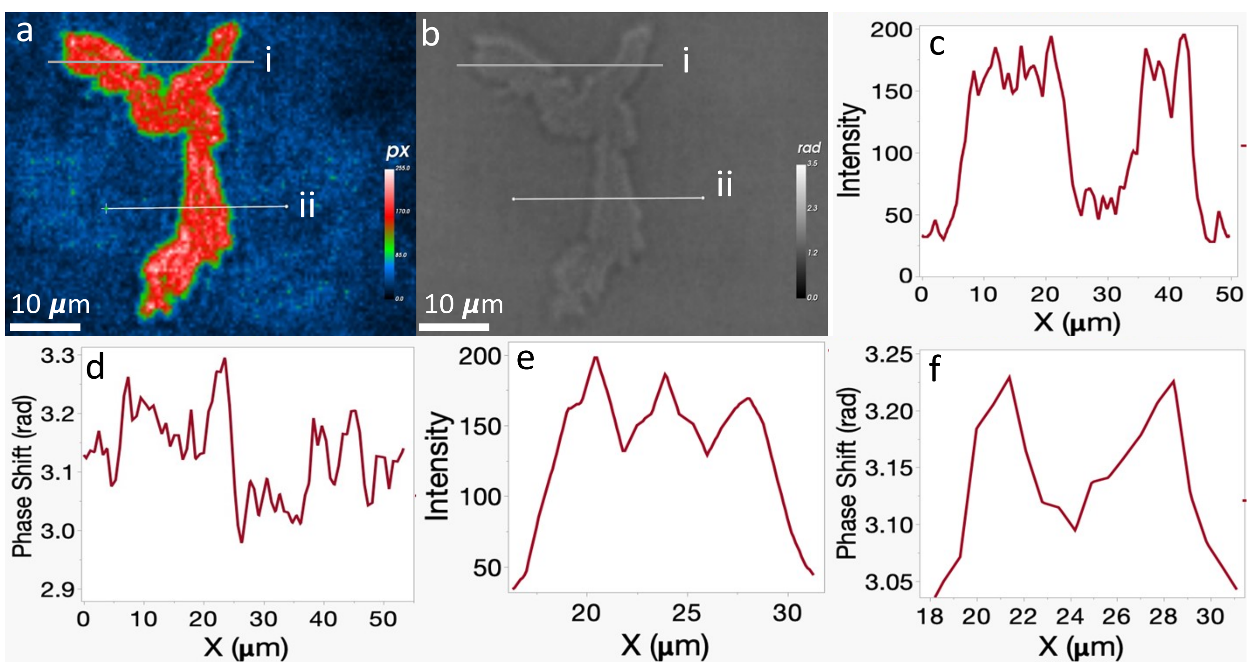
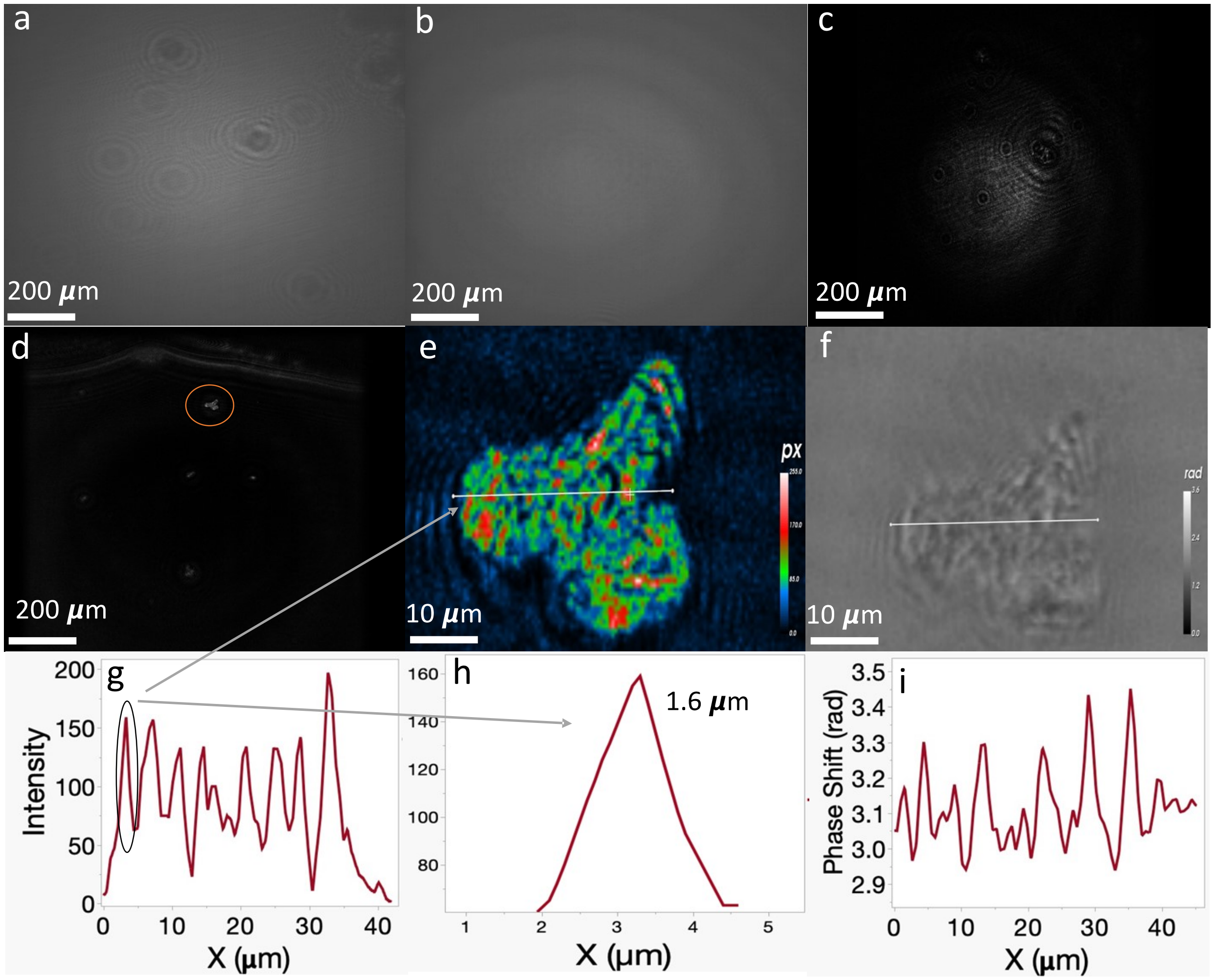


Figure S3. Holographic reconstruction of graphite solid particles. (a) intensity reconstruction of graphite at z = 3429 μm, (b) phase reconstruction of graphite at z = 3456 μm, (c-e) The intensity profiles of graphite particles across the particle crosscuts (i) and (ii) respectively, (d-f) Phase profiles of graphite particles across the particle crosscuts (i) and (ii) respectively.



*1.6 *μm*: shows the size of the first particle of graphite referred by the grey arrow.

Figure S4. Reconstruction of the intensity information for BC particles in the solid phase. (a) Raw hologram recorded for solid BC particles, (b) background hologram recorded without BC particles, (c) contrast hologram obtained after subtracting the background hologram from the raw hologram, (d) contrast hologram with particle in focus from (c) at z = 1403 μm, (e) Intensity reconstruction of BC particles, (f) Phase reconstruction of BC particles. The horizontal lines in e and f show the crosscut across the particles, g, h, i, (g) The intensity profile of BC particles across the particle crosscut, (h) crosscut of the first particle in the agglomerate which showed the smallest size, (i) The phase profile of the BC particle crosscut.

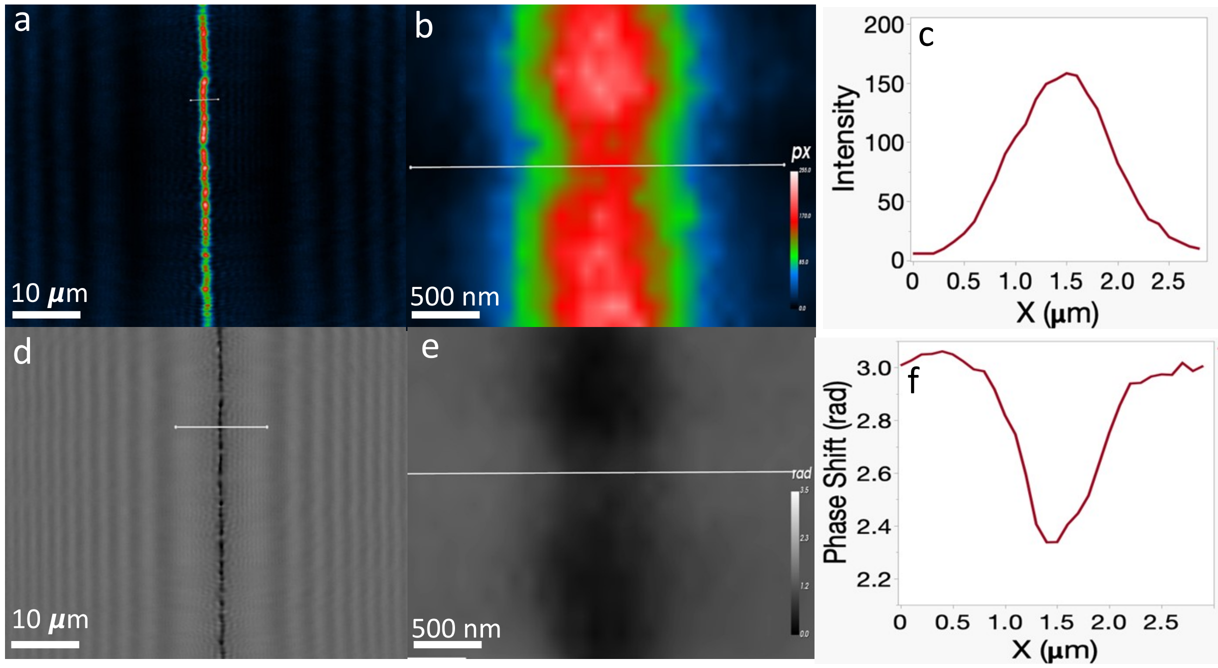


Figure S5. Holographic reconstruction of single walled carbon nanotube solid particles CNT. (a) intensity reconstruction of single walled CNT at z=396 µm, (b) zoomed in area of (a), (c) The intensity profile of single walled CNT particles across the particle crosscut, (d) phase reconstruction of single walled CNT at z=414 µm, (e) zoomed in area of (d), (f) Phase profile of single walled CNT particles across the particle crosscut.

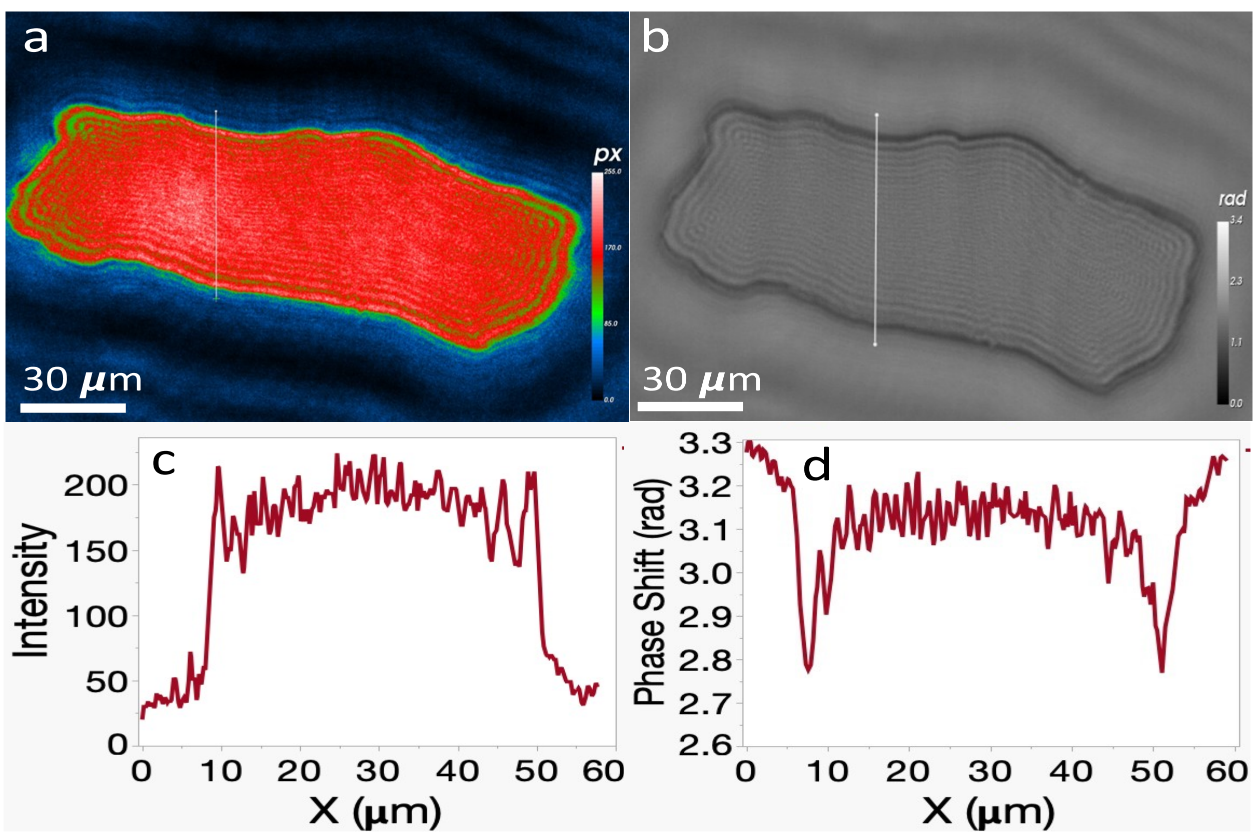


Figure S6. Holographic reconstruction of single walled carbon nanotube solid particle agglomerates. (a) intensity reconstruction of single walled CNT at z = 396 µm, (b) phase reconstruction of single walled CNT at z = 414 µm, (c) The intensity profile of single walled CNT particles across the particle crosscut, (d) Phase profile of single walled CNT particles across the particle crosscut.

Supplementary Tables:

Table S1: Different aerosol characterization methods with their advantages and limitations.

Type		Method	Advantages	Limitations	Reference
Physical Analysis	**Online**	Differential Mobility Analyzers DMAs: Scanning Mobility Particle Sizer (SMPS), Differential Mobility Particle Sizer (DMPS)	-Classify aerosols based on electron mobility. -Provide information about the particles surfaces and volume. -Used to measure smaller particles. (2-1000nm)	-No real-time information about shape, morphology, and refractive index	(23, 24)
		Optical Analyzers: Optical Particle Sizer (OPS) Optical Particle Counter (OPC)	-Determination of particle size and volume from intensity of scattered light. -Used for larger particles (300-100000nm)	-No real-time information about shape, morphology, and refractive index.	(23, 24)
	Offline	Filter Sample Gravimetric Techniques.	-Use of impactors for collection of aerosols on filters. -Gives information about mass concentration, shape, and surface.	-Possibility of adsorption/desorption of water. -Particle loss during air sampling period. -Operating cost	(23, 24)
Chemical Analysis	**Offline**	Impactors/Factors	-Use of impactors for collection of aerosols on filters which is followed by chemical characterization using techniques (HPLC, GC, IC and HNMR)	-Long sampling periods where most atmospheric changes are very fast (short term). -Possibility of contamination or decomposition of compounds	(23, 24)

		Aerosol Mass Spectrometer (AMS)	-Measurement of size and chemical mass for non-refractory constitutes (ammonium, total nitrate, total organics…) -Used for submicron aerosols.	-Limited to a number of atmospheric chemical species.	(23, 24)
		Photoacoustic Extinctionometer (PAX)	-Measurement of concentration of black carbon in air. -Higher time resolution than offline methods.	-Doesn't give information about morphology, size and phase	(23, 25, 26)
	Online	Scanning/Transmission Electron Microscopy (S/TEM) and Atomic Force Microscopy (AFM)	-Gives information about size, phase, morphology. -2D characterization of aerosols.	-Only work in stationary mode. -No 3D information about aerosols.	(23, 25, 27, 48)
		Fourier Ptycography and Optical Diffraction Tomography	-2D and 3D characterization of aerosols.	-Only work in stationary mode. -No in-situ and real time information.	(23, 25, 28, 29)
		Nano Digital In-line Holography Microscope (Nano-DIHM)	-Real-time and in-situ characterization of aerosols in 2D and 3D. -Determination of physicochemical characteristics and surface properties of aerosols in real-time and in-situ. -Provides 4D trajectories of aerosols in dynamic mode. (3D +time) -Works in both stationary and dynamic mode.	-Limited to submicron particles. -Difficulty in achieving spatial resolution.	(23, 25)