Years of results regarding secondary organic aerosols reduce uncertainty in climate projections

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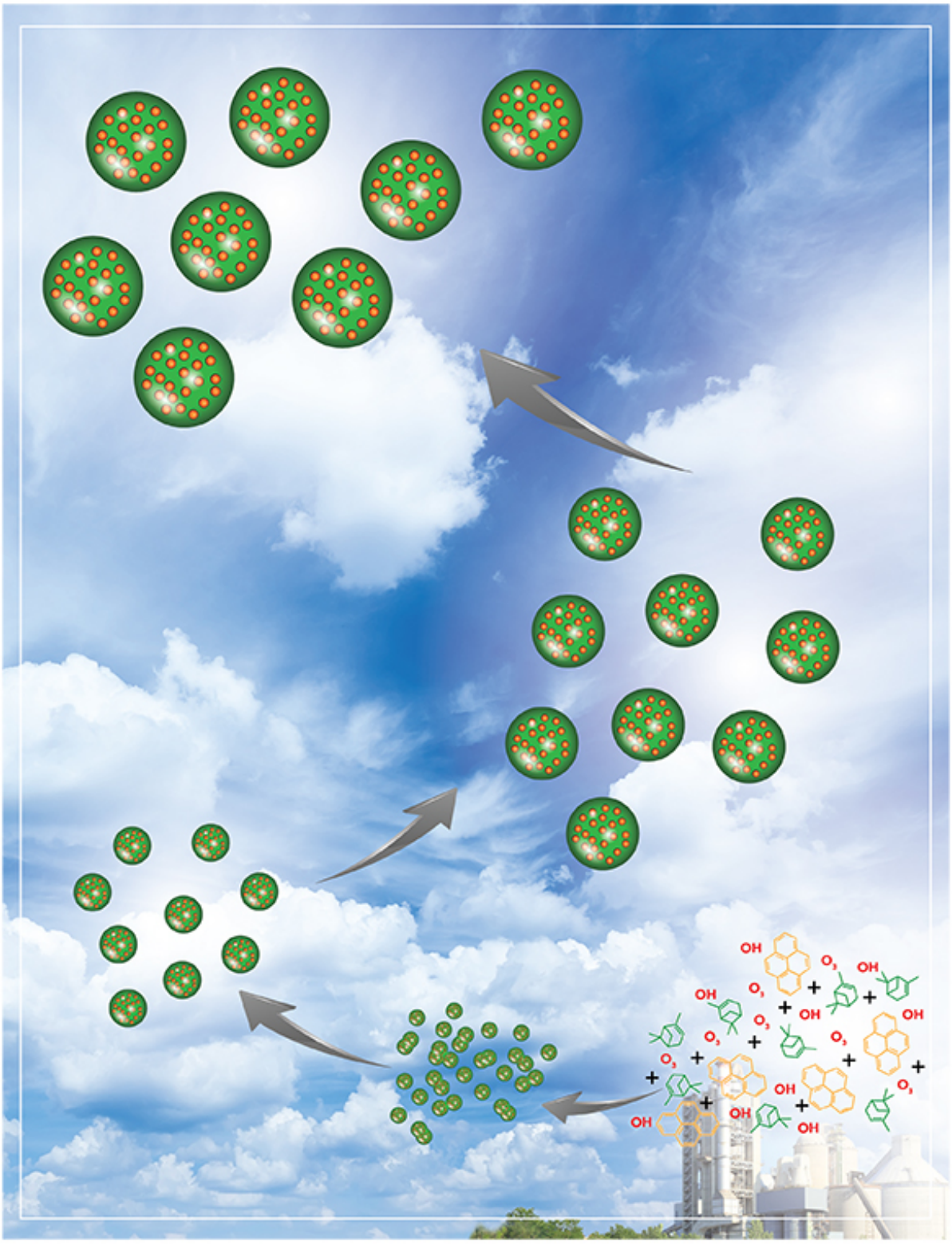


Contrary to long-standing assumptions, our laboratory and field data show that

SOA particles are highly viscous semi-solids, trap volatile organic molecules during formation, evaporate orders of magnitude slower than assumed, and, therefore, are not at equilibrium with the gas phase.

For the past 20 years, a large portion of the particles measured in the atmosphere were missing from models. At best, models were able to explain one-tenth of the carbon-rich secondary organic aerosols, or SOA, measured in the air. The problem turned out to be a series of fundamental assumptions used in the models due to a lack of experimental data. The models assumed the particles were liquids that responded to changes in the surrounding atmosphere by rapid condensation or evaporation and in-particle mixing. The models predicted the particles, especially the small ones, were rather short-lived on their journey away from their sources. All of these assumptions and more were proven false by Dr. Alla Zelenyuk and her colleagues at Pacific Northwest National Laboratory, Imre Consulting, University of Washington, and University of California at Irvine.

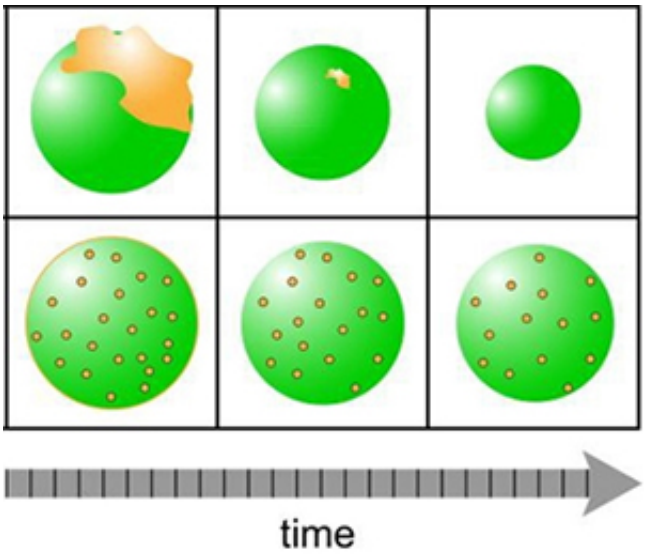
These particles generated by natural and anthropogenic sources are of interest because of their effect on air quality and health, and their impact on climate by scattering and absorbing solar radiation and influencing the formation and properties of clouds.

Seeking to find the reason that most of the particles found in the atmosphere are missing from the models, Zelenyuk and her colleagues began by designing novel approaches to investigate what these particles really look like and how they evolve in atmosphere. They built SPLAT II, a second generation single particle mass spectrometer. Highly precise and sensitive, this instrument allows users to study basic processes and properties of nanoparticles. Usually located in DOE's EMSL, a national scientific user facility, the instrument also travels to field sites, working

on the ground and aboard research aircraft. SPLAT II has been flown to many sites in the United States, South Korea, and Germany.

Using SPLAT II, the researchers measured the shape, size, density, composition, evaporation rates, and many other relevant properties of SOA-containing particles in laboratory and field. The results show that SOA particles are far from the perfect liquid spheres assumed in models. The actual SOA particles are highly viscous semi-solids that have complex compositions and multifaceted structures.

The team was the first to directly measure chemical diffusivity of tracer molecules in SOA particles, determining that these particles are tar-like, and millions of times more viscous than assumed. This viscous nature allows the particles to trap toxic polycyclic aromatic hydrocarbons, PAHs, and other chemicals that would otherwise quickly evaporate. Zelenyuk and her colleagues discovered a symbiotic relationship between the PAHs and the SOA particles. The PAHs hitchhike along and, in the process, help the SOA survive longer. This hitchhiking phenomenon explains how anthropogenic pollutants from California freeways and biomass burning in Asia can be transported far away from their sources to the pristine environments, such as the Arctic.



When airborne particles (green) form before pollutants known as PAHs (yellow) glob on, the particles evaporate quickly (top row). But when the particles form in the presence of PAHs, which is what likely happens in nature, the long-lasting particles can take the pollutants for a long-distance ride (bottom).

SOA particles, even without PAHs, are long lasting. Some scientists believed the particles evaporated nearly instantaneously at higher levels of humidity. Zelenyuk and her team found, however, that the particles evaporate orders of magnitude slower under all atmospherically relevant conditions.

Now, Zelenyuk and her team are working with atmospheric modelers to add this information, independently verified, into models. The team created a new, measurement-based paradigm of SOA that now underlies three-dimensional regional and global climate models to properly represent SOA properties. These models yield significantly higher loadings and longer atmospheric lifetimes, thus reconciling the long-standing discrepancy between observations and model predictions.

More information: "Experimental determination of chemical diffusion within secondary organic aerosol particles." *Physical Chemistry Chemical Physics* 15 (8), 2983-2991 (2013). DOI: 10.1039/C2CP44013J

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