ROBERT J. CHARLSON 1936–2021

Robert (Bob) Charlson, a pioneer in understanding the complex roles in climate of atmospheric aerosol particles, died in Seattle on 28 September 2021. Bob invented new instruments to measure atmospheric aerosol particles, led the first studies quantifying the radiative forcing by anthropogenic aerosols, and developed a hypothesis for how ocean biological processes may stabilize Earth's climate system.

Charlson was a faculty member at the University of Washington since 1965, beginning his faculty career in civil engineering and later moving to the Department of Atmospheric Sciences. Bob was born in San Jose, California, and received B.S. and M.S. degrees in chemistry from Stanford University, and a Ph.D. from the department in 1964 under the supervision of Konrad Büttner. Bob then spent a year as a Fulbright scholar in the United Kingdom with B.J. Mason at Imperial College before returning to Seattle.

It is hard to overstate the impact that Bob's research has had on the field of aerosol science and on the atmospheric sciences more broadly. He was a leader in demonstrating essential linkages in biological, geophysical, and chemical phenomena. Bob was an inventor of multiple instruments to measure atmospheric aerosols, garnering him six patents. The nephelometer, which measures light scattering by aerosols, resulted in the first patent to bear royalties for the University of Washington. It is still in broad use today around the globe. The nephelometer work established that atmospheric aerosol particles have characteristic sizes and associated light-scattering properties, which ran counter to the scientific thinking at the time. He was also involved in organizing the first large-scale, integrated study of aerosols, particles, gases, meteorology, and photochemistry in the early 1970s. Bob and his graduate students were subsequently involved in many such studies around that globe that have been fundamental to our understanding of the role of aerosols in both air quality and climate. To this day, the nephelometer is broadly used in such field campaigns and in routine monitoring of atmospheric aerosols.

As his many colleagues and graduate students could attest, Bob was a man of many ideas, and he generously shared these ideas with others. His interdisciplinary interests led him to collaborate with scientists across the biological, chemical, and geosciences, and his infectious enthusiasm and drive brought many different institutions and individuals together.

Observations that Bob made with colleagues using the nephelometer in the 1960s and early 1970s provided observational evidence that established the scattering of light by aerosols as a potentially important component of Earth's radiation budget and hence of climate and climate change. In the mid-1980s, Bob organized a meeting at Schloss Ringberg in the Bavarian Alps, which led to recognition that light scattering by sulfate aerosols, now known as the direct effect, might be as important as the indirect (cloud) effect that Sean Twomey had shown could exert a significant climate forcing. This led to the initial calculation of scattering by sulfate in a global transport model by Bob and his collaborators at Stockholm University. Bob led the first paper quantifying the global effects of anthropogenic aerosol pollution on the climate through direct interaction with sunlight. The Ringberg meeting also drove Bob to champion a paper that would summarize current knowledge about forcing by sulfate aerosols, including indirect effects on clouds, and outline what was needed to better evaluate this effect. That paper, published in 1992 in *Science* magazine, brought together broad input and expertise from universities and multiple federal agencies. Although almost 30 years old, this paper continues to be cited 100 times each year.

Bob similarly conducted foundational work on the impacts of natural and anthropogenic emissions as a source of cloud condensation nuclei. Another of Bob's inventions, the Counterflow Virtual Impactor (CVI), collects cloud droplets and ice crystals allowing the extraction of their chemical components in situ for greater understanding of hydrometeor origin and evolution. Early CVI work by Bob and his students measured the light-absorbing material inside cloud droplets and established that the chemical composition of cloud droplets is size-dependent. Bob was fascinated by low supersaturation conditions found in polluted environments, where in extreme cases the sharp boundary between (subsaturated) haze and (supersaturated) clouds is blurred. Bob's 1997 *Nature* paper with Patrick Chuang and John Seinfeld at Caltech showed that, under such conditions, extreme competition for water vapor and kinetic limitations on the growth of haze particles can lead to clouds where a significant fraction of droplets is not

strate that two stable regimes for aerosol concentration can exist, with each dominated by a different aerosol sink mechanism. This work demonstrated for the first time that collision-coalescence of droplets leading to the formation of drizzle can exert a profound impact on aerosol concentrations in maritime air masses. Over time, this has led to the understanding that the macrophysical and microphysical properties of clouds are often strongly interdependent.

Perhaps Bob's most well-known paper, now cited more than 3,000 times, focused on the potential for climate stabilization through a natural feedback loop involving the production of dimethyl sulfide (DMS) by ocean phytoplankton. What is now known as the CLAW hypothesis (after the last names of the contributing authors, Charlson, Jim Lovelock, Andi Andreae, and Steve Warren), originated as the result of a serendipitous constellation that brought Lovelock, Andreae, and Charlson together at the University of Washington (UW) in October 1984. At the time, Lovelock was looking for a real-world mechanism that would represent the climate stabilization mechanism that he had

activated. This work, although requiring some revision, inspired a critical examination of the assumptions used to treat droplet activation (in a 2001 paper in *Tellus* led by Thanos Nenes) and helped to establish highly accurate parameterizations of cloud droplet activation used in climate models today. Another 1999 *Nature* paper, led by Maria Christina Faccini, showed that surface-active organic compounds in aerosols may lower surface tension and facilitate cloud droplet formation. How this and other "chemical effects" on droplet formation were reshaping the science of cloud droplet formation was then synthesized in a 2001 paper led by Bob in *Science*.

Bob's work on the importance of aerosol composition on cloud droplet activation stimulated Jost Heintzenberg and colleagues to design and develop the Leipzig Aerosol Cloud Interaction Simulator (LACIS) to study the formation and growth of cloud droplets under near-atmospheric supersaturations. LACIS can reproduce the thermodynamic conditions of atmospheric clouds as realistically as possible and continues to produce transformative science. Bob was fascinated by the entire aerosol life cycle and provided inspiration for the 1990 Nature paper led by Marcia Baker that developed a simple model to elegantly demon-

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formulated as Daisyworld. Charlson had worked on the powerful climate effects of aerosol particles acting as cloud condensation nuclei, and Andreae had proposed DMS as the primary natural source of atmospheric sulfate aerosol and thus the dominant source of natural marine CCN. After seeing a presentation by Charlson on the topic at UW, Warren provided a quantitative assessment of how changes in the clouds from DMS would affect sunlight. The CLAW paper hypothesized that an increase in DMS production would increase the sunlight reflected by clouds and serve as a cooling effect on climate. Although the subsequent effect of this cooling on DMS production was left as a question for further research, the CLAW paper spurred much new work that generated a wealth of new understanding about the importance of aerosol–cloud interactions in the climate system.

Over many years, Bob built strong ties between UW and Stockholm University, generating many productive collaborations. These led to the first regional estimate of sulfate aerosol cooling and the first estimate of global climate forcing by sulfate aerosols based on a global model of the sulfur cycle. Together with Henning Rodhe, Bob showed that the pH in rainwater even in remote areas is significantly lower than expected from equilibrium with CO2. Collaborations with Stockholm University led to the invention of the counterflow virtual impactor, a unique instrument to collect and analyze the properties of aerosols within cloud droplets. In addition, two books resulted from the UW–SU cooperation: *The Legacy of Svante Arrhenius: Understanding the Greenhouse Effect and Earth System Science (1998); From Biogeochemical Cycles to Global Change (2000)*.

Bob's pioneering work established numerous ways in which aerosol processes impact the climate system and showed that anthropogenic aerosol emissions can drive climate change. His work helped inspire a generation of scientists and helped justify NASA's investments in spaceborne backscatter lidar, which began with a technological demonstration on the Space Shuttle in 1994 and culminated in the 2006 launch of the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) instrument on the *CALIPSO* satellite. Bob contributed important insights to studies using CALIOP that led to observational estimates of aerosol direct radiative forcing from smoke layers above clouds, and a realization that roughly 50% of low-lying clouds over the global oceans are transparent to lidar.

Bob mentored 35 Ph.D. and eight M.S. students at UW across his career and is remembered as a supportive and generous advisor. He served as informal mentor to many other young scientists, providing encouragement to think about big problems without making them seem intractable. Bob officially retired from UW in 1999 and then accepted an appointment by the king of Sweden to be his professor of environmental science for the year 1999–2000. He was a contributing author on the 4th Assessment of the Intergovernmental Panel on Climate Change that won the 2007 Nobel Peace Prize, and continued to work with collaborators at UW and around the world until shortly before his death.

Bob began his career with instrumentation to make local limited measurements, steadily broadening his interests and contributions over the years to focus on global biogeochemical systems, a unique trajectory. In all his work, Bob displayed an ingenious ability to capture the essence of highly complex problems and reduce them to the essential simple equations that could then be used to quantitatively express key chemical and physical connections between aerosol, clouds, radiation, and climate.

Bob is survived by his wife, Pat; his children, Daniel (wife, Maureen) and Amanda; nieces Kathryn and Erika; and eight grandchildren. Bob will be missed by the many students he mentored, his colleagues, friends, and family.

—Robert Wood