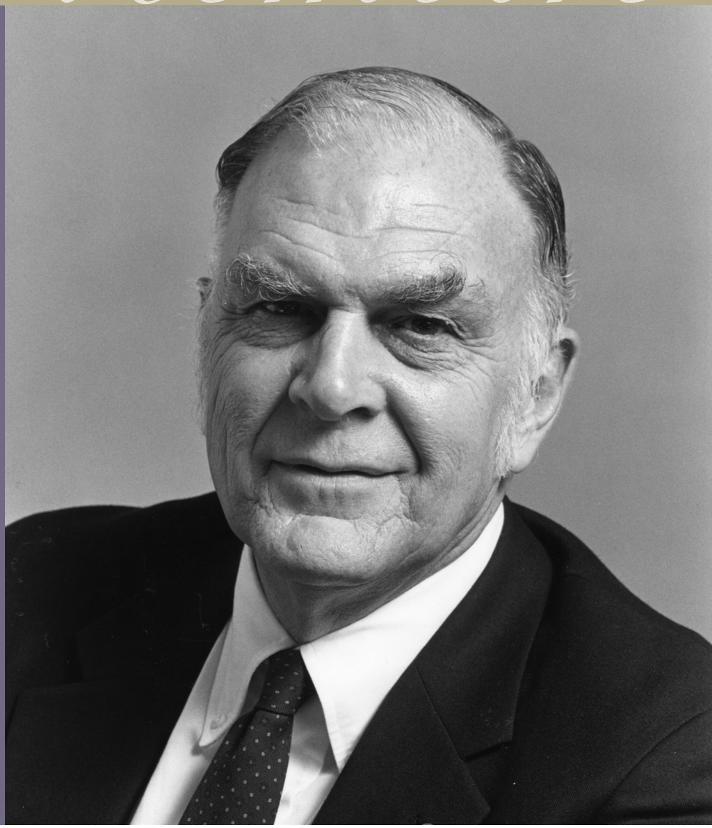
F. Sherwood Rowland

1927–2012

BIOGRAPHICAL *Memoirs*



A Biographical Memoir by
B. J. Finlayson-Pitts,
D. R. Blake and
A. R. "Ravi" Ravishankara

NATIONAL ACADEMY OF SCIENCES

FRANK SHERWOOD **ROWLAND**

June 28, 1927–March 10, 2012

Elected to the NAS, 1978

By B. J. Finlayson-Pitts, D. R. Blake, and A. R. "Ravi" Ravishankara

F. S. Rowland, known to all as "Sherry," belongs to the elite club of Nobel laureates (1995) for his proposal in 1974 that chlorofluorocarbons (CFCs) would be transported into the upper atmosphere. He recognized that the harsh ultraviolet (UV) light occurring at higher altitudes would photolyze CFCs to release highly reactive halogen atoms, which very efficiently destroy thousands of ozone molecules for each CFC molecule in a chain destruction process via homogeneous gas phase catalysis involving Cl atoms. Stratospheric ozone is the naturally formed UV shield for the Earth's surface. Sherry and postdoctoral fellow Mario Molina suggested that such a depletion of the ozone layer would decrease UV filtering, thereby negatively affecting life on Earth as we know it.

So, was receiving the Nobel Prize in Chemistry the most exciting moment of Sherry's career? When asked this question a few years later, his somewhat surprising response was "no." It was actually when the truth and enormity of his research findings hit home—that "Aha!" moment that comes rarely in a scientific career but makes all the hard work worthwhile.

That was quintessential Sherry: a focused, dedicated scientist whose curiosity constantly drove him to search for that "Aha!" moment in new areas. In that vein, his research ranged from hot atom chemistry to CFC chemistry and photochemistry, air pollution, greenhouse gas sources, and human breath analysis as markers of disease and drug efficacy. But in addition to that (some would say more than that), Sherry was at the leading edge of a group of scientists who were translating their science into public policy forums covering a wide range of atmospheric science issues. These included air pollution, climate change and stratospheric ozone depletion—three of the most profound environmental issues of our times. His willingness to communicate clearly the implications of research results to a broad spectrum of stakeholders, and with urgency, was met at that time with considerable skepticism from much of the chemistry community. This

Some of Sherry's outreach to legislators, including Vice President Gore, and President Clinton, as well as Prime Minister Thatcher. He is with his wife, Joan.

was particularly so when it involved problems that significantly affected human health and welfare. But, as he said in 1997 at a White House roundtable, "… if you believe that you have found something that can affect the environment, isn't it your responsibility to do something about it, enough so that action actually takes place?" This sounds much easier than it was at the time. He became *persona non grata* for some years. Invitations to give seminars dried up for almost a decade, and graduate students and postdoc applicants were rare, as he negotiated a minefield of industrial and political opposition. At one point, it was even suggested that he was a KGB agent!

Young scientists today take it as a given that they must communicate and interact broadly, from "Brews and Brains" informal talks for the general public to presentations that help educate policymakers, legislators, and regulators regarding the implications of their work. Sherry's factual, persistent, and dogged efforts to highlight the long-term dangers posed by CFCs are seen now as an ideal to be emulated by younger generations.

Early Life and Education

Sherry was born in Delaware, Ohio, on June 28, 1927. He had a solidly middle-class upbringing, his father being chair of the Department of Mathematics at Ohio Wesleyan University. Portending his future, Sherry was academically accomplished, starting high school at age 12. He was active in sports from an early age, being on the tennis team at age 13 in high school. At 16, he was too young for military service in World War II when he graduated high school, so he went straight into the undergraduate program at Ohio Wesleyan, where he played varsity baseball and basketball. When he turned 18, he entered the Navy as a trainee radar operator. Never having traveled outside of Ohio, he requested that he be discharged in California so he could see another part of the United States. From there, he hitchhiked back home and returned to Ohio Wesleyan to finish his degree in 1948, with a triple major in chemistry, physics, and mathematics.

He was accepted into the Ph.D. program at the University of Chicago, his parents' alma mater, where he joined the research group of Willard Libby, who was doing hot atom chemistry. Libby (who had earlier worked with Harold Urey, another Nobel laureate, during the Manhattan Project) was to later earn a Nobel Prize in 1960 for his work on developing the ^{14}C dating method. For his doctoral research with Libby, Sherry studied the chemistry induced by the recoil of bromine when it undergoes an (n,2n) reaction to transform to a new isotope with enormous energies that were more than enough to break chemical bonds. His first paper with Libby is a classic example of clarity and simplicity in writing. This paper also gives away the secret long held from the atmospheric chemistry community: that Sherry's first name was "Frank." It was actually something of a family tradition to ignore first names!

Sherry playing minor league baseball.

Unsurprisingly, Sherry continued to be active in athletics while a graduate student. At the University of Chicago, Sherry was first baseman and clean-up batter for three years. During that time, he also played for a minor league baseball team in Oshawa, Ontario, in Canada and, taking over as manager, led them to the semi-pro championship. Needless to say, this time away from the lab was not fully appreciated by his Ph.D. advisor, who toward the end of his degree, told Sherry he could be successful if he was not so "lazy"! With his height and imposing physique, Sherry later was famous for participating in pick-up basketball games during scientific meetings and surprising his colleagues with his athletic abilities.

While at Chicago, Sherry met Joan Lundberg, who was his soul mate from the beginning, and they married on June 7, 1952. Theirs was a special partnership that lasted for six decades until his passing in 2012. He relied on Joan throughout that time for her wisdom and advice…and as it turns out, recording measurements in the lab during his Ph.D. research while he was refereeing basketball games to earn some extra income. He made no secret of her influence, later recognizing her as the honorary co-recipient of his Nobel Prize. A unique insight into their relationship came during his testimony to the Subcommittee on Environmental Pollution of the Committee on Environment and Public Works of the United States Senate on June 10 and 11, 1986 regarding CFCs:

Senator Chafee: "Suppose you were king and you had what you might call unlimited authority. What would you do about this problem?"

Dr. Rowland: "If I were king, the first thing I would do is consult with the queen, who is sitting behind me and who has a very good view on what the sensible things to do in such cases are."

For the rest of their time with the committee, Joan was addressed by the legislators as "The Queen"!

Early Career

On being awarded his Ph.D., Sherry obtained a position in 1952 as an instructor at Princeton University. During his time there, he spent summers at Brookhaven National Laboratory on Long Island, New York, carrying out research on hot atom chemistry and radiation chemistry. But he came to find that Princeton did not afford him the independence that he desired. For example, when Sherry successfully obtained his first research grant, the department chair told Sherry that he could not have a single-investigator grant and that he (the chair) should be included. When an opportunity came to move to the University of Kansas, Sherry and Joan jumped at it, joining the Department of Chemistry in 1956. Over the next eight years, Sherry established an internationally recognized research group on hot atom and radiation chemistry. He had access to some of the important sources of neutrons and other radiation sources, including the new cyclotron and a very large 30 Curie Po^{210} neutron source to carry out his studies.

Starting after his arrival in Kansas, the first hundred or so of Sherry's papers were all about hot atom and radiation chemistry, far-ranging in their breadth and interpretations. Some of his students and postdocs from this period later became well-known photochemists and chemical physicists. Using radioactive isotopes, Sherry elucidated some of the intricacies of chemical reactions, such as exploring the anti-Markovnikov reactions. He studied the mechanisms of various chemical reactions to show how radicals react. Indeed, his curiosity with hot atom chemistry extended to fluorine atom reactions, including the conversion of F^{19} to O^{18} following the (n,2n) reaction. So, long before his venture into the CFC saga, Sherry was fully aware of the strength of the C-F bond and why the CFCs would be very stable. Even during this period, Sherry was already looking at the composition of the atmosphere. For example, he measured the ratio of the hydrogen gas HT to water vapor (HTO) in the atmosphere (T is tritium, a very unstable isotope of hydrogen with two neutrons). The hot atom research also established Sherry as an essential scientist for the Atomic Energy Commission (AEC), later the Department of Energy (DOE). Indeed, some his earliest studies on CFCs were funded by the DOE.

One of the authors of this memoir (ARR) got into atmospheric chemistry because he heard Sherry's talk on CFCs as a graduate student. He attended Sherry's lecture thinking he would be talking about hot atom chemistry and came out of the lecture a new person, wanting to do atmospheric chemistry.

Sherry mentored a string of amazing chemists who went on to brilliant careers. Examples include John Root, Burdon Musgrave, E. K. C. Lee, and others. During this time, Sherry also navigated towards using the rapidly developing gas chromatography (GC) method, which stayed with him for the rest of his scientific career studying atmospheric chemistry. Until his passing and indeed continuing to the present time, Sherry's laboratory has been full of GC instruments!

University of California, Irvine, and CFC Research

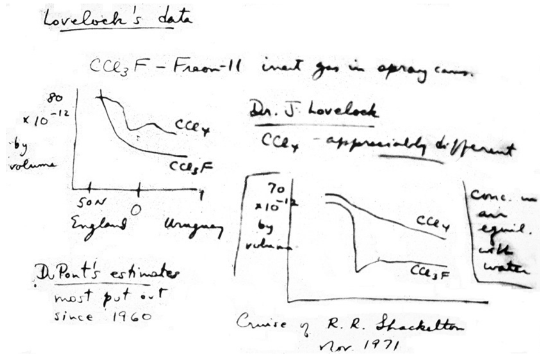
The University of California, Irvine (UCI) was being developed in the early 1960s, and in 1964 Sherry and Joan moved to California, where he took up his role as founding chair of the Department of Chemistry. There was little there at the time but open fields and cattle. Temporary buildings served as offices while research and teaching facilities were being designed and built. One of the first was the Physical Sciences building, which was renamed Rowland Hall in 1996. This included a nuclear reactor for Sherry's hot atom work in the basement of the building.

Sherry looking into the reactor core at UCI.

During his time as chair, Sherry was responsible for eleven new hires and, most importantly, setting the tone that would provide the underpinnings for the department as it developed over the years. To this day, the chemistry department is characterized by a collegial, cooperative "rising tide lifts all boats" attitude, rather than one of internal competition. Even after winning the Nobel Prize, Sherry continued to interact with and support the department in many ways that might not be expected from someone with constant demands on his time and attention.

Sherry was an early supporter of women in science, long before the importance of inclusion was recognized. He mentored many female graduate students and postdoctoral fellows, and one of his early faculty hires as department chair was Marjorie Caserio, at a time when female faculty were few and far between. Decades later, he supported hiring one of the authors (BJFP) when women were still not well represented in chemistry departments at R1 universities.

It was at UCI that Sherry began his most important work on CFCs, after his retirement as department chair in 1970. During a train ride from Salzburg to Vienna after the 1971 International Atomic Energy Agency, he had a conversation with a meteorologist who had also been at the conference. The meteorologist mentioned to Sherry that he saw a great need for meteorologists and chemists to work together to address unsolved problems, which intrigued Sherry. He invited Sherry to an Atomic Energy Commission meeting of meteorologists in Florida the following year, and Sherry enthusiastically accepted. At this 1972 meeting, James Lovelock's CFC measurements were presented. Sherry was a prodigious note-taker, scribbling throughout all seminars and meetings that he attended. He saved all of these notes, and some from the meeting involving Lovelock's data were presented are shown in the photo.



Some of Sherry's notes from 1972 AEC meeting.

The CFC story is an interesting example of questioning accepted wisdom and seeing things beforehand that seem obvious in hindsight. Sherry had been funded by the AEC for many years during his hot atom work. In the early 1970s, CCl_3F, CCl_4, and CH_3I had been detected at parts per trillion (ppt or 10^{-12}) levels in the air by James Lovelock. Lovelock had developed the electron capture detector that was widely used in gas chromatography, and it was exquisitely sensitive to halogenated organic compounds. Because CCl_3F and CCl_4 lack reactive C-H bonds, they are inert in the lower atmosphere, so much so that CFCs were thought to be inert tracers of air masses. Indeed, CFCs are used as tracers of ocean movements even to this day. Rowland and postdoc Mario Molina realized that these very same inert chemicals would rise to the stratosphere, blow apart in the environment of harsh ultraviolet light, and destroy the fragile ozone layer.

At this time, atmospheric chemistry was just in its beginning stages, emerging largely out of research on air pollution in the troposphere (the first 15 kilometers or so above the Earth's surface), which became severe with increased fossil fuel combustion especially after World War II. It was only in 1970 that the hydroxyl (OH) radical was proposed as a major atmospheric oxidant. Studies of the kinetics and mechanisms of its reactions, along with those of O_3, were in their infancy. Even less was known about the chemistry of the stratosphere (15-50 kilometers above the Earth's surface).

Sydney Chapman had identified natural cycles involving the formation and destruction of ozone in the stratosphere in the 1930s, and Hampson had suggested the catalytic role of the natural OH radicals in reducing ozone amounts. Relatively little further attention was focused on stratospheric processes until 1971, when Paul Crutzen showed that nitrogen oxides (called NO_x, the sum of NO + NO_2) could also play a significant role in determining stratospheric ozone levels. This finding was almost simultaneously augmented by Harold Johnston's suggestion that NO from supersonic aircraft would deplete the ozone layer. Indeed, the role of NO_x was thought to be the major issue for ozone layer depletion. To this situation, Rowland and Molina would add research detailing the harmful effects of the CFCs, which in the end became the most critical issue.

Molina had earned his Ph.D. from the University of California, Berkeley around this time and came to join Sherry's group as a postdoctoral fellow. Sherry gave Mario the choice of several different possible research projects, one of which was tracing the fates of CFCs in the atmosphere. Given the "accepted wisdom" that they were unreactive, this might have been considered something of a fool's errand. It was, however, in line with Sherry's advice to the next generations of scientists: "Don't look under the light. Go out into the darkness." Mario chose to work on this project, and very quickly, he and Sherry concluded that although CFCs were unreactive in the troposphere, this was not the case in the stratosphere. Ozone and oxygen shield the lower atmosphere from UV, but at higher altitudes, the filtering drops off so that the CFCs are exposed to wavelengths of light they can absorb, decomposing to generate highly reactive chlorine atoms that very efficiently destroy ozone in a chain reaction:

$$Cl + O_3 \rightarrow ClO + O_2 \qquad (1)$$

$$ClO + O \rightarrow Cl + O_2 \qquad (2)$$

$$\text{Net: } O_3 + O \rightarrow 2\,O_2$$

Fortunately, the rate coefficients for these reactions were known from the laboratory work of Michael Clyne and his students at St. Mary's College in London. In December 1973, Sherry and Mario had checked and double-checked their calculations and concluded there were no errors that would affect their conclusions. Joan would ask Sherry each day how the work was going. Upon developing confidence in their calculations, Sherry's response was: "Good, but it might be the end of the world." Reduction in ozone with an associated increase in UV reaching the Earth's surface was predicted to have a wide range of deleterious impacts, such as increased skin cancer and cataracts, and devastating effects on materials and biological systems, including agriculture.

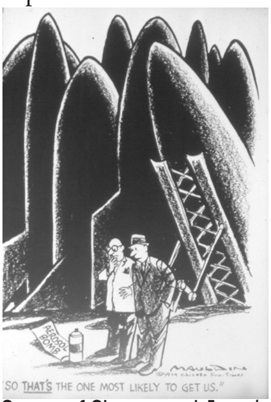
The work was submitted to *Nature* in January 1974. Quoting from that paper:

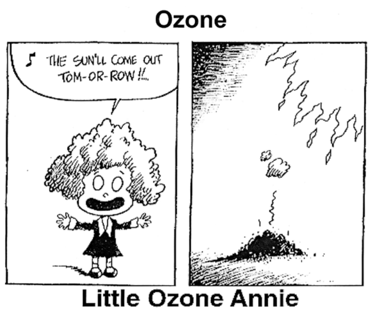
> *"Photodissociation of the chlorofluoromethanes in the stratosphere produces significant amounts of chlorine atoms, and leads to the destruction of atmospheric ozone….It seems quite clear that the atmosphere has only a finite capacity for absorbing Cl atoms produced in the stratosphere and that important consequences may result."*

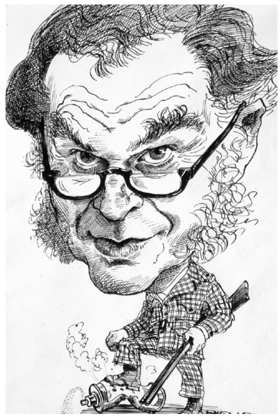
Interestingly, Rich Stolarski and Ralph Cicerone had suggested such a cycle at a 1973 conference in Canada that was published in the conference proceedings in the *Canadian Journal of Chemistry* in 1974. They were investigating the possible impacts of the expected frequent Space Shuttle launches. Stolarski and Cicerone had proposed that chlorine from the Space Shuttle program (it was supposed to use ammonium perchlorate) could destroy the ozone in the stratosphere. At the time, however, the rockets were thought unlikely to be a significant source of chlorine to the stratosphere. When Stolarski and Cicerone became aware of the Molina-Rowland *Nature* paper identifying CFCs as a chlorine atom source, they added a note to the proof of their manuscript, and this work was cited as a seminal contribution to the 1995 Nobel Prize awarded to Molina, Rowland, and Crutzen for their research on the role of chlorofluorocarbons and nitrogen oxides in the destruction of stratospheric ozone. Thus began a lifelong close friendship between Sherry and Ralph.

The following decade saw an explosion of research in atmospheric chemistry. Analytical capabilities as well as the development and application of satellite technology and sampling platforms that could reach the stratosphere provided increasing support for the CFC ozone-destruction hypothesis. This was supported by a National Academy of Sciences report in 1976. But definitive evidence of decreasing stratospheric ozone in mid-latitudes was challenging owing to the plethora of factors that provide variability in natural ozone levels. These factors included the predictable solar cycles, spatial variability, and short-time meteorological variability. The predicted mid-latitude ozone depletions were supposed to be in the range of 5 percent to 15 percent by 2050 in a scenario with continued emissions of ozone-depleting gases that would be tens of times larger than in the 1970s. It was only after the publication of the *Nature* paper by Farman et al. in May 1985, which reported the dramatic annual austral springtime loss of ozone over Antarctica (the "ozone hole"), that the issue was raised in the consciousnesses of the world and its leaders. Intense field campaigns to probe the origin of the ozone hole irrefutably confirmed the original idea set forth in the 1974 Rowland-Molina paper, but with an additional "wrinkle." In the Antarctic situation, the very dramatic declines in ozone were shown by Susan Solomon and Sherry to result from the rapid recycling of temporary halogen reservoirs, such as chlorine nitrate, back to photochemically active halogens on the surface of polar stratospheric clouds common in the very cold Antarctic winter. The upshot of these amazing advances made it clear that ozone layer depletion presented a clear and present danger.

Based on the testimony of Rowland, Molina, Cicerone, and others, the U.S. Congress had already banned the use of CFCs as spray agents in aerosol cans in 1978. Meanwhile, broader regulations awaited the concrete evidence demonstrated by the ozone hole. This ultimately led to the 1985 Vienna Convention and the 1987 Montreal Protocol on Substances that Deplete the Ozone Layer, which has been amended and adjusted several times as new science has been carried out. As the stratospheric ozone story developed over a decade or more, there were many cartoons published about CFCs and ozone destruction. Sherry and Joan purchased the originals of many of these, and a wonderful set of framed copies still adorns the walls of their home. Some of their favorites are shown here.







Some of Sherry and Joan's favorite cartoons.

Sherry was always very interested in clear communication of the science underlying societally significant environmental issues. For example, during a meeting to assess the status of the ozone layer held in Les Diablerets, Switzerland, Sherry started wondering how best to communicate such information clearly. A discussion between Sherry, Susan Solomon, and several others quickly turned to a practical mode for such communications. Thus was born the now familiar "Common questions about the ozone layer," co-authored by Susan Solomon and Sherry Rowland. This document has morphed over the years and helped pave the way for the effective communications that are now common in scientific assessments.

One of the authors of this memoir (DRB) joined Sherry's group in 1978 as a Ph.D. student. The central role in climate change of increasing levels of CO_2 from fossil fuel combustion was well known, but the extent to which other greenhouse gases, such as methane, were contributing was not as well recognized. Sherry suggested that Don quantify methane in air samples that were being collected for CFC analysis. In 1978, methane was known to be a component of the atmosphere but thought to be present at a constant concentration. Rowland and his students were one of the first groups to report, starting in 1978, increasing concentrations of methane, which continues today and provides the longest global methane record. As the years went by, the number of gases quantified in the air samples increased, as did the group's involvement in atmospheric sciences campaigns around the world. The associated photos show Sherry collecting a surface sample, aboard the NASA DC-8 aircraft during an East Asian airborne campaign in 1991, and in the lab with DRB.

Luck is where preparation meets opportunity. In 1991 Sherry was at a meeting in Mexico City and happened to be eating lunch with the mayor of Mexico City. The mayor mentioned that Mexico City had bad air quality and asked if Sherry could get involved in helping them better understand the causes of pollution. An undergraduate student was sent from UCI to Mexico City during the Thanksgiving break, and she collected eight air samples at various locations. She also collected two air samples from the stoves in two homes. The major component of the stove fuel was expected to be methane but it turned out to be liquefied petroleum gas (LPG), which in Mexico City was comprised of about 50 percent propane, 40 percent butanes, and 10 percent highly reactive butenes. Interestingly, when the ambient samples were analyzed, they looked almost identical to the stove gas. A year later, a paper was published indicating that if the LPG contained less of the very reactive butenes, the ambient air would have less ozone-forming potential. Two years later, Mexico City passed a law requiring LPG to contain no more than 1 percent butenes, and air quality improved.

In another new venture, Sherry teamed with medical scientists to measure trace organic gases in human breath as a marker for disease and drug efficacy. The use of breath samples to determine health conditions was not as easy as they thought. But by replacing CFCs with HFC-134a, a substitute that is now used as the propellant in asthma inhalers, Sherry and his group were able to estimate how much of the prescription medications within the inhalers was actually getting into the lungs of the users. It was found that subjects with poor lung function were not getting all of the steroids into their lungs and thus were being underdosed.

Given that the field of atmospheric chemistry was not widely recognized until late in the twentieth century, scientists contributing to this area came from many other disciplines and fundamental sciences. A less tangible but lasting impact of Sherry's work has been the emphasis on a broad view of atmospheric processes and the need to bring expertise from many different fields to bear on complex environmental problems. This belief was the impetus for his leadership in founding the Department of Earth System Science at UCI, with Ralph Cicerone as its first chair. Taking a global view of issues that affect the Earth and its inhabitants, as well as the strategy of developing the fundamental science and then translating results to be usable for public policy, has gained strength in the last several decades, thanks to leadership and resolve by recognized scientists like Sherry.

Honors and Personal Life

In addition to service to the department and UCI campus, Sherry contributed to many professional organizations. This included being Foreign Secretary of the U.S. National Academy of Sciences (1994-2002), Founding Co-chair of the Inter-Academy Panel on International Issues (1995-2000), and President of the American Association for the Advancement of Science (1992).

Needless to say, Sherry earned many honors and awards in addition to the Nobel Prize. These included being elected a member of the American Academy of Arts and Sciences, the National Academy of Sciences, the Institute of Medicine, the American Philosophical Society, and the Royal Society (U.K., Foreign Member). Amongst other accolades, he was awarded the Roger Revelle Medal of the American Geophysical Union (1994), the Silver Medal of the Royal Institute of Chemistry (U.K., 1989), the Tyler World Prize for Environmental Achievement (1983) for which he later served on the board until his passing, the Japan Prize in Environmental Science and Technology (1989), the Peter Debye Award in Physical Chemistry (1993) from the American Chemical Society, the Esselen Award (1987), the Environmental Science and Technology Award (1983) and the Tolman Medal (1976). He also received 18 honorary degrees, including from his alma maters Ohio Wesleyan University and the University of Chicago.

Finally, as a person, Sherry was without fail intellectually honest, of the highest integrity, and generous. He did not accept industrial support or consulting projects to avoid even the perception of bias. When he gave seminars and talks, he would not say "I did this…," but rather "X person did this, and Y person in my group did that." The ultimate example of this was insisting from the very beginning of their CFC and stratospheric ozone discovery that Mario, a postdoctoral fellow in his lab at the time, share equal credit, all the way to the Nobel Prize.

The morning that the Nobel Prize was announced, there were, of course, many congratulations. But as one of the authors (DRB) of this memoir noted:

> *"I wasn't the only one (in tears)….I saw other faculty members in tears that day. That shows you what kind of friend and colleague Sherry Rowland has been. People would have come and shook his hand even if he had been a jerk, but there would not have been that joy."*

When something amused Sherry, he would cock one of his eyebrows, and a grin would spread over his face. This photo is the closest we came to capturing that impish expression.

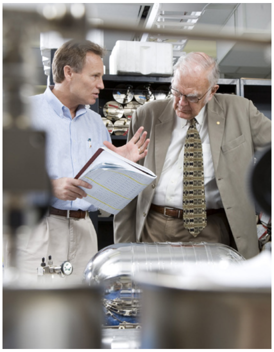
Sherry and Joan were devoted opera fans, taking advantage of their world travels to attend operas in many locations. Their favorite opera companies were the Metropolitan Opera in New York, the Salzburg Opera in Austria, and the Santa Fe Opera in New Mexico, which they attended during summers for many years. Joan bought Sherry a formal opera cape, which he wore to many performances.

Sherry was a tall person (6' 5") and was even more imposing in the cape. One evening as they were walking back to their hotel from the Metropolitan Opera, a homeless man looked up and, seeing Sherry, said in astonishment, "Wow, there goes Count Dracula in a suit!" Sherry and Joan chuckled about that for many years thereafter.

Joan and Sherry in his opera cape.

Sherry was a devoted family man. Dinner at home with the family and time spent with Joan, their daughter Ingrid, and son Jeff was sacrosanct. He was equally proud of his granddaughters, Taylor and Lindsey, and daughter-in-law Kristi.

Sherry passed away peacefully at home on March 10, 2012, from complications of Parkinson's disease. His passing was noted not only in the world of science, but on nightly news broadcasts in the United States and in a number of other countries. This shows the importance of his work and the acknowledgement of his role in saving the world. Additionally, 2018 documentary, *Saving Planet Earth: Fixing a Hole*, traces the progress from the 1974 *Nature* paper to the Montreal Protocol and provides some detailed and interesting perspectives on the process and how it unfolded. The Montreal Protocol and its amendments represent a remarkable achievement in global cooperation, setting the stage for developing similar approaches to address climate change, another area to which Sherry has contributed significantly.



Photos of Sherry out sampling and in his UC Irvine lab with Don Blake.

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