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A Giant Among Chemists

Nobel Laureate F. Sherwood Rowland discusses his almost six-decade-long career as researcher and public policy advocate

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IN MANY WAYS, Nobel Laureate in Chemistry <u>F. Sherwood Rowland</u> is an imposing figure: He is imposing in his 6'5" height, in the breadth of his intellect, in his ability to work in cross-disciplinary areas, and in his willingness to step outside the confines of academia and effect change in the policy arena.



William J. Cooper/UC Irvine

Rowland

Rowland, 80, was awarded the Nobel Prize in Chemistry in 1995, along with <u>Mario J. Molina</u> and <u>Paul J. Crutzen</u>, for pioneering research on how chlorofluorocarbons destroy stratospheric ozone. By 1987, this work had led to the signing of the Montreal Protocol on Substances That Deplete the Ozone Layer, widely considered the most successful international environmental treaty ever.

Last month, C&EN interviewed Rowland in his office at the University of California, Irvine, where he is the Bren Research Professor of Chemistry & Earth System Science.

Since the time Frank Sherwood Rowland was born in the small central Ohio town of Delaware in 1927, he completed everything early. He started first grade at age five, skipped fourth grade, entered high school at 12. He enrolled at Ohio Wesleyan in 1943 at the age of 16. During those war years, there were only 30 to 40 civilian males on campus, along with 200 naval officer trainees and 1,000 women, Rowland says. After two years of college, he enlisted in a Navy program to train radar operators. He never saw combat because the war with Japan ended while he was still in training

In 1946, Rowland returned to Ohio Wesleyan and completed a B.A. two years later with a triple major in chemistry, physics, and mathematics. He then began graduate studies at the University of Chicago, where his parents had earned degrees many years earlier. There, Willard F. Libby, who would win the 1960 Nobel Prize in Chemistry for his invention of carbon-14 dating, was randomly assigned by the chemistry department to be Rowland's mentor. Rowland settled happily into Libby's research group and did his Ph.D. work on radioactive atoms.

"Almost everything I learned about how to be a research scientist came from listening to and observing Bill Libby," Rowland says. Although he encouraged intensive critical thought and hard work on experiments, Libby gave his students a great deal of leeway about how they used their time. Rowland's thesis topic involved the chemical state of cyclotron-produced radioactive bromine atoms. "It was an unbelievably exciting time in the physical sciences at the University of Chicago," Rowland observes. In 1942, physicist Enrico Fermi had built the first nuclear reactor on a squash court under the football stands. After the war, many leading scientists from the Manhattan Project began teaching in the chemistry and physics departments at the university.

During those college years, athletic activities were almost as important to Rowland as science was. At Ohio Wesleyan, he played on the baseball and basketball teams and—with so few civilian men on campus—wrote much of the sports section for the university newspaper. In the Navy, he devoted considerable time to playing basketball and left the service as a noncommissioned officer with a rating of Specialist (A), 3rd Class, with "A" for athletics.

At the University of Chicago, he received a multiyear Atomic Energy Commission (AEC) national fellowship—the forerunner of the current National Science Foundation national fellowships—but was best known as a varsity baseball and basketball player. The university was highly unusual in that it permitted graduate students to compete in intercollegiate athletics. Rowland also spent two summers on a Canadian semiprofessional baseball team, playing for and even managing the team when it won the Canadian championship in 1950.

Shortly after his 25th birthday in 1953, Rowland finished his Ph.D. thesis and married Joan Lundberg, also a graduate of the university. He then accepted a position as an instructor in chemistry at Princeton University. During his four years at Princeton, he spent summers working in the chemistry department at Brookhaven National Laboratory on Long Island.

At Brookhaven in 1954, Rowland began to do research that turned heads. "I took a mortar and pestle and ground up lithium carbonate with glucose and put this mixture in the neutron flux of a nuclear reactor. The neutrons reacted with lithium-6 to produce tritium atoms with 2.7 million eV of recoil energy, more than enough to carry them into the neighboring glucose particles," he explains. "About 15% of the tritium ended up bonded directly to carbon in glucose. Because glucose was just one example of a molecule with carbon-hydrogen bonds, that experiment made it apparent that this probably would happen with many molecules with carbon-hydrogen bonds. This research ended up as an article in *Science*."

That work, in such a high-profile publication, helped Rowland win a national competition for research support run by AEC. "So I went back to my chairman at Princeton and told him about the AEC offer. He refused to sign such a proposal, saying 'No. You are too young.' " Rowland was 27 at the time.

THE FOLLOWING YEAR, 1956, when the University of Kansas offered Rowland a position as assistant professor, the Princeton chairman changed his mind, telling Rowland he could accept AEC funding. "But I hadn't changed my mind. I went to the University of Kansas," Rowland says. "The day I arrived in Lawrence was the first day of the AEC contract, and it lasted 38 years."

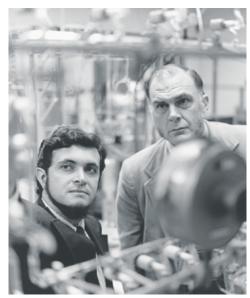
At the time, the University of Kansas had just completed a new chemistry building with special facilities for radiochemistry. A number of graduate students interested in radiochemistry joined Rowland's research group that summer. Other students joined as well, including several from China and Japan. Most of the research focused on chemical reactions of energetic tritium atoms. Rowland received tenure after two years at Kansas, and he advanced to

professor in 1963.

In 1964, when the University of California, Irvine, was in the planning stages and had no students or buildings, Rowlanc accepted a position as chair of its future chemistry department. "In 1964, if you looked out the window, there was no green anywhere, only brown ranch fields. The city of Irvine had not yet been built," he recalls. "My first year, 1964-65, was a year of starting a chemistry department and recruiting six additional faculty including Robert W. Taft from Penn State and Marjorie Caserio from a senior research position at Caltech."

Every few years at Irvine, Rowland tried to bring variety into his group's research by taking on some new, challenging aspect of chemistry. First, he extended the work into radioactive tracer photochemistry, using tritium and carbon-14. Then, he expanded the the research portfolio to include chlorine and fluorine chemistry, using the radioactive isotopes chlorine-38 and fluorine-18. "I deliberately followed a policy of trying to instill some freshness into our research efforts," he says.

In 1970, when Rowland retired from the department chairmanship, his research efforts took a more abrupt turn. The firs Earth Day occurred in 1970, and "the state of the environment had become a significant topic for discussion by the public and within my family," he says. He had begun to think about how radioactivity might be used to study the environment. One pivotal action, he recalls, was to attend a meeting in Salzburg, Austria, entitled "Environmental Applications of Radioactivity," sponsored by the International Atomic Energy Agency.



UC Irvine

Laureates Rowland (right) and Molina in 1974.

"After the conference, on the way from Salzburg to Vienna, I shared a train compartment with William Marlow, an AEC program manager," Rowland says. "I was talking with him about carbon-14 dating because I had worked with Nobel Laureate Libby. I found out that Marlow was organizing conferences that brought together chemists and meteorologists AEC felt the two fields needed some cross-fertilization.

"Marlow invited me to attend a conference in Fort Lauderdale, Fla., in 1972," Rowland continues. "One of the first lecturers, the late Lester Machta, reported on results obtained by James Lovelock, a British scientist who had invented a highly sensitive way to measure trace gases. Machta, former director of the National Oceanic & Atmospheric Administration's Air Resources Laboratory, said that Lovelock had detected small amounts of trichlorofluoromethane [CFC-11, or Freon] in the atmosphere and that the gas could be of no conceivable harm in the atmosphere." From his measurements, Lovelock had also calculated that the amount of CFC-11 in the atmosphere almost equaled the total ever produced.

Although CFCs are inert in the lower troposphere, Rowland realized that they could be broken down by short-wavelength ultraviolet radiation in the stratosphere-eight to 30 miles aboveground. "When I wrote my next yearly research proposal to AEC, I added one page, saying I would like to look into what eventually becomes of CFC molecules in the atmosphere," Rowland recalls. At the time, almost 1 million tons of CFCs—considered chemically iner and harmless—were used each year in household aerosol sprays, refrigeration systems, and certain industrial processes.

The most important, and in some respects the most difficult, event in Rowland's research career was the discovery shortly after preparing this AEC proposal that CFCs can destroy large amounts of stratospheric ozone.

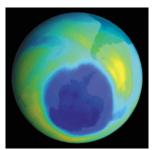
Over three months from October through December of 1973, Rowland and Molina, who had recently joined Rowland's group as a postdoctoral fellow, used data from a variety of published studies and calculated that CFC molecules released near the surface of the planet would, over a period of decades, end up in the stratosphere. There, UV radiation would break off chlorine atoms. Each chlorine atom would react immediately with one molecule of ozone and set in motion a chain reaction that would end up destroying thousands of ozone molecules. Rowland and Molina estimated that if CFC production continued at its then current rate, 7-13% of the ozone layer would be lost. But if CFC production kept increasing at its then-current growth rate of 10% a year, the losses would be far greater than that. Because the ozone layer protects life at Earth's surface from the sun's dangerous UV radiation, the researchers knew theirs would be no run-of-the mill reporting of new data.

"When we realized there was a very effective chain reaction, that changed the CFC investigation from an interesting scientific problem to one that had major environmental consequences," he says. When Rowland saw what the calculations meant-that the continuing destruction of stratospheric ozone could eventually kill most plants and animalshe was aghast. "You don't get many chills down your back when you look at scientific results," he explains, but this was clearly one of those moments.

Rowland and Molina tried to find flaws in their calculations but could find none. In January 1974, they wrote up their findings and submitted a paper to *Nature*. After having heard nothing from the journal in five months, Rowland called the office and learned that the paper had been lost temporarily but had been found and was working its way through the review process. One problem the manuscript faced, he says, was that it crossed disciplinary boundaries. Chemists knew little about the atmosphere so they were reluctant to review it, and meteorologists knew little about chemistry, so most refused to comment.

Rowland and Molina's paper was published that June, and their results were replicated a few weeks later by several scientists, including Crutzen, then a meteorologist working with the National Center for Atmospheric Research in Boulder, Colo.

The science behind the CFC-stratosphere ozone connection was looking solid and the stakes were so high that the research triggered behavioral and then policy responses. In 1976, a National Academy of Sciences study and congressional hearings on CFCs finally attracted public attention, and Americans started shunning aerosol cans with CFC propellants. In the same year, the U.S. announced a formal ban on nonessential uses of CFC aerosol propellants. However, CFC aerosol propellants were still used in Europe and Japan, and CFCs were still employed as refrigerants throughout the world.



NASA

Blue Hole Deep blue indicates stratospheric ozone depletion over Antarctica in September 2001.

For his part, Rowland spoke outwardly about the implications of ozone destruction at public and scientific meetings. The CFC industry, government officials, and even some academic chemists, however, attacked the ozone destruction theory and Rowland's reputation. They argued that the theory of ozone destruction was based on calculations, not actual measurements in the stratosphere, and demanded much stronger evidence that ozone was indeed being destroyed. Each time Rowland lectured, CFC industry coalitions released statements disputing his claims, he recalls. The number of invitations he received to speak at universities declined.

JUST THE SORT of evidence his critics demanded finally came to light. The Halley Bay Station of the British Antarctic Survey had been taking direct ground-based measurements of total ozone for decades. But the machine—a Dobson UV spectrometer—had been programmed to discard measurements that appeared to be outliers, either unreasonably high or low. Consequently, it was rejecting very low ozone data when producing the final monthly output. In 1984, Joseph C. Farman and his colleagues at BAS decided to look at the raw data and found that stratospheric ozone had

actually decreased greatly since the 1960s. In 1985, they published an article in Nature announcing that stratospheric ozone was reduced 40% over Antarctica in late September, the month when ozone is most likely to be highly depleted in the south polar region. Their findings were confirmed in 1986 by Nimbus satellite data.

These findings galvanized international action. In 1987, 56 countries agreed under what became known as the Montrea protocol to 50% cuts in CFC use and production. And in 1988, DuPont announced that it would stop making CFCs. In subsequent years, the protocol was strengthened to require an eventual worldwide phaseout in the production of CFCs and other ozone depleters.

"It has been a very successful protocol," Rowland says. "One of the things about working in the atmosphere is that it is pretty hard to hide what has been done. So if there were black-market production of molecules released to the atmosphere, they would be seen not just by us but by other groups."

Now 20 years later, Rowland continues to cull secrets from the atmosphere and to find novel ways of thinking about the atmosphere. Currently, he and <u>Donald R. Blake</u>, also a chemistry professor at UC Irvine, codirect a research group. Half the members study the outer atmosphere and the other half study the "inner atmosphere," the chemical constituents in the breath of individuals.

"We are continuing our measurements of the composition of the atmosphere, which were started in 1978," Rowland says. "We take measurements all over the Pacific from Beryl, Alaska, to the southern tip of New Zealand and 70 to 80 places in between."

The Rowland-Blake group is measuring about 150 different compounds, including many chlorine and bromine species, from both aircraft and ground stations. "We are focusing on methane in particular because it has been the number two greenhouse gas over the past two centuries," Rowland says. Indeed, methane is behaving strangely. Before 1980, its concentrations rose steadily in a fairly straight line, he notes. Since 1980, the increase has been less uniform and progressing more slowly.

Rowland theorizes that for economic reasons, many gas companies around the world capped most of their leaks in the 1980s and early 1990s and this slowed the rise in methane levels. Now, whenever the concentration of methane rises, ethane levels also rise, he explains. The only source of methane that simultaneously produces ethane is biomass burning, he says. "By elimination, the measurements pretty much say that biomass burning is a source of pulses in both methane and ethane," he says. In 1997, for example, large fires in Indonesia produced significant amounts of both gases.

In addition to the remote atmosphere, "we have also measured trace gases in cities around the world," Rowland says. "We were surprised to find some of the highest concentrations of hydrocarbons in Oklahoma City, which was not on ou list of well-known smog producers," he explains. It is polluted from poorly capped old oil and gas wells and from oil and gas storage facilities. "The very highest concentration that we've found was in Odessa, Texas," a center of the oil and gas industry with many old fields, he points out.

And then there is breath itself. In collaboration with experts from the UC Irvine Medical School, members of the Rowland-Blake group are also investigating the inner atmosphere. They are trying to answer the question: "What does each individual bring to the room in exhaled breath and what does that tell you about him or her?" Rowland says. They are using the same methods to analyze trace chemicals in breath that they use to study the atmosphere.

The group has completed two major studies, one of cystic fibrosis patients and another of diabetic patients. The atmosphere contains an amount of carbonyl sulfide (OCS) on the order of 600 ppt. The Rowland-Blake group found the if healthy individuals breathe in air with 600 ppt of OCS, their exhaled air has about 350 ppt. In other words, the process of breathing removes about 250 ppt of OCS from the air. But individuals with cystic fibrosis exhale air with an average OCS concentration of 490 ppt-an average net uptake of only 110 ppt.

The study suggests that the more severe the cystic fibrosis, the higher the concentration of OCS in the exhaled air, Rowland says. It is known that bacteria produce OCS. "The breath levels of OCS might be elevated in cystic fibrosis patients because of increased presence of bacteria in their lungs," he explains. When further developed, the method might be used as a marker of bacterial colonization in lungs of patients with cystic fibrosis, he notes.

THE ROWLAND-BLAKE group has also been measuring methyl nitrate levels in the exhaled breath of diabetics and healthy control patients. As the sugar content of the blood rises, the methyl nitrate content of the exhaled breath goes up, Rowland explains. A breath test could perhaps be used eventually as a surrogate test for blood sugar, he says. It

would be much less invasive than drawing blood.

Even as he treads new research ground, Rowland continues his efforts to influence U.S. and international policies. Now, as chair of the Committee on International Affairs at the <u>National Academy of Sciences</u> and as cochair of the <u>Interacademy Panel on International Issues</u>, he is well-positioned to make his views known about U.S. science and environmental policies.

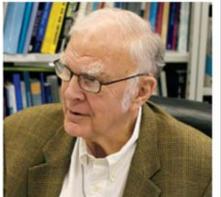
In 2004, for example, he and more than 60 other prominent scientists signed a statement charging the Bush Administration with undermining the integrity of science in policy decisions. The statement says: "President George W. Bush and his staff have systematically distorted scientific information to further policy goals in health, environment, and nuclear weaponry."

Also, in January of this year, Rowland spoke of the need for 17 new Earth-observing satellite missions over the next decade. "As soon as NASA announced manned travel to the moon and Mars as goals, people recognized that that's where the money would be going and there wasn't likely to be enough to fully fund a third priority-Earth-observing systems," he says.

It is very important to maintain continuity in satellite observations, Rowland notes, because if one satellite stops measuring a particular parameter and another begins gathering similar data a few years later, it is hard to intercalibrate between measurements.

Throughout his academic career, interactions with students have been very important and satisfying to Rowland. Now, after many years of success in conducting research and in influencing environmental policies, Rowland spends a fair amount of time traveling to campuses and talking with chemistry majors. "There are questions students want to ask me not because of general interest in some aspect of chemistry, but because they are interested in my personal interaction with an area of chemistry at the time the work was being done," he says. "The question of what it is like to do scientific research comes up regularly." The fact that Rowland is 80 and has no plans to retire indicates that to him it's a very satisfying way to spend his time.







William J. Cooper/UC Irvine

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