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Paul J. Crutzen
Hans Günter Brauch *Editors*

Paul J. Crutzen: A Pioneer on Atmospheric Chemistry and Climate Change in the Anthropocene

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To Terttu

Ms. Terttu Crutzen; Prof. Dr. Paul J. Crutzen and Minister Doris Ahnen, Rhineland-Palatinate Minister for Education, Research, Further Education and Culture during the symposium “The Anthropocene” on 2 December 2013 in Mainz on the occasion of his 80th birthday. Photo by Carsten Costard

Prof. Dr. Paul J. Crutzen received the Order of Merit of the land Rhineland-Palatinate from Doris Ahnen, Rhineland-Palatinate Minister for Education, Research, Further Education and Culture, during the symposium “The Anthropocene” on 2 December 2013 in Mainz. Photo by Carsten Costard

Foreword

Paul Crutzen grew up near the center of Amsterdam, the Netherlands, during a period of economic depression and World War II. Clearly, he was not born with a silver spoon in his mouth. However, his family held up and made the best of it, using simple means to overcome the difficult period. This included playing soccer in the streets and having arithmetic and language contests between father and son. Actually young Paul excelled in soccer, languages as well as mathematics, but could not develop these talents in full, at least not immediately. In such an environment practical solutions are needed, and he took up an education in civil engineering to design bridges in his water-rich country. This was a good career start because later Paul showed that bridges can also be built between scientific disciplines and communities.

With the love of his wife from Finland, Terttu Soininen, Sweden became their country of choice to reside and raise two wonderful daughters. It was a lucky decision, because the University of Stockholm offered Paul the opportunity to advance one of his talents, mathematics, which he pursued within a practical context: meteorology. Another fortunate coincidence was that he became involved in computer modeling early on. In those days, he was debugging the code for predicting weather and storms by listening to the hums of the mechanical processors. The development of these skills was instrumental for an unparalleled vocation in atmospheric and environmental science.

During his graduate studies, Paul addressed the middle and upper atmosphere, which initially seemed to be of academic interest only. However, he became aware of the great importance of stratospheric ozone and in particular the risks of ozone depletion. Although not trained as a chemist, he found that some of the reactions used to model the stratospheric fate of nitrogen oxides and their impact on ozone were severely misrepresented as they could not explain the observations of ozone.

By adopting more realistic reaction rate constants he discovered that nitrogen oxides from human-made sources could actually damage the ozone layer, which was absolutely unexpected. It was published in Paul's third first-author paper "The influence of nitrogen oxides on the atmospheric ozone content," for which he would be awarded the Nobel Prize 25 years later.

By combining models of meteorology and ozone, Paul pioneered the field of atmospheric chemistry, and showed how local emissions can have a global effect, even though the substances in question occur in minute, i.e., trace amounts. With his work, that has had an impact well beyond his own field, he followed in the footsteps of pioneers in chemistry in the past centuries such as Scheele, Priestley, Lavoisier, and Laplace. Like Paul, they were also intrigued by the chemical composition of air, what controls it, and tried to unravel its importance for life on Earth. The central role of nitrogen oxides in stratospheric ozone chemistry was the first of Paul's impressive series of discoveries.

Prof. Dr. Jos Lelieveld opening the Symposium on 2 December 2013 in Mainz on the occasion of Prof. Paul J. Crutzen's 80th birthday devoted to The Anthropocene. The photo was taken by Carsten Costard

After his theoretical studies in Stockholm, Paul spent some years at Oxford University to work with experimental data and enhance his proficiency in chemistry. During this period an international discussion sparked about the environmental consequences of high-flying supersonic aircraft, such as the Concorde, of which entire fleets were planned. Since these aircraft release their nitrogen oxide containing exhausts directly into the ozone layer, Paul and other scientists engaged in a debate about the risks. Suddenly, the formerly academic research landed in the center of attention. This became the story of Paul's life, as many of his discoveries inspired both scientific and public discussion. He does not shy away from expressing his opinion, also with an unwelcome message. Fortunately, the fleet of high-flying aircraft was limited to a few Concordes—of which the last were decommissioned in 2003.

Paul continued to make major contributions to stratospheric chemistry. For example, he explained how nitric acid clouds cause the Antarctic ozone hole. At the same time, he also turned his attention to the troposphere, which is the air layer that connects with the biosphere and where weather and climate take place. The troposphere is also prone to air pollution, while it is cleaned by oxidation reactions. The self-cleaning capacity relies on the presence of reactive hydroxyl radicals that convert pollutant gases into more soluble compounds that are removed by rain. The primary formation of hydroxyl radicals in turn is from ozone. While most ozone is located in the stratosphere, protecting life on Earth against harmful ultraviolet radiation from the Sun, a small amount is needed in the troposphere to support the self-cleaning capacity. While previous theories had assumed that tropospheric ozone originates in the stratosphere, Paul discovered that much of it is actually chemically formed within the troposphere. The formation mechanism is similar to the creation of ozone pollution in photochemical "smog".

Prof. Dr. Jos Lelieveld opening the Symposium on 2 December 2013 in Mainz on the occasion of Prof. Paul J. Crutzen's 80th birthday devoted to "The Anthropocene". The photo was taken by Carsten Costard

In the mid-1970s, Paul and his family moved to the USA at the invitation of renowned research institutions in Boulder, Colorado. He helped establish stratospheric research programs, though mostly pursued tropospheric chemistry, and also investigated large-scale sources of air pollution. One such source is biomass burning, which had previously gone unnoticed because it mostly takes place in scarcely populated regions. Paul found that it is actually a major source worldwide, especially in the tropics. This was confirmed by field measurement campaigns, and later by satellite measurements. The effects are particularly large in the southern hemisphere where other human-related pollution sources are smaller than in the strongly industrialized northern hemisphere.

The next major step in the life of the Crutzen's was the move to Mainz, Germany, when Paul accepted a directorship at the Max Planck Institute for Chemistry where he succeeded another pioneer in atmospheric chemistry, Christian Junge. In Mainz, Paul became involved in scientific as well as political discussions about the impacts of using nuclear weapons through his seminal paper "The atmosphere after a nuclear war: twilight at noon." It built on knowledge gained from the biomass burning studies. A main effect that was expected from nuclear conflict is that giant smoke plumes from large-scale fires submerge the world into darkness, similar to some of the super volcano eruptions or asteroid impacts that changed the fate of the Earth in prehistoric times.

In Mainz he also developed the next generation of comprehensive meteorology–chemistry models to simulate the biogeochemical cycles of reactive nitrogen and carbon compounds, for example methane, and their control of tropospheric ozone and the self-cleaning capacity. His group created the first numerical tool of this kind, ultimately leading to the development of atmospheric chemistry-climate and Earth system models. In this period, I first met Paul, which was a milestone in my career and personal life. He bestowed me the honor of writing a preface in my book "Air pollution in the troposphere" (in Dutch), and I am delighted to now continue the tradition. Subsequently I started a Ph.D. project under Paul's guidance, to study the role of clouds in tropospheric chemistry. In the past three decades we developed a close personal friendship, which includes our wives Terttu and Tineke.

In my pre-Mainz life, I was involved in aircraft measurements, and Paul had turned me into a computer modeler, or perhaps something in-between. Actually we learned that this is a useful combination and a good basis for collaboration, also after my career continued elsewhere. One example is the "Indian Ocean Experiment," planned with the climate change pioneer V. Ramanathan (Ram). We performed measurements with several instrumented aircraft, ships, and satellites and combined the results with computer modeling. This provided indisputable evidence of the environmental impacts of an atmospheric brown cloud with the dimension of several million square kilometers. This huge pollution haze was shown to not only affect air quality on a large scale, but also influence the monsoon and climate in South Asia. Later, several more brown clouds were identified, for example in East Asia.

Paul's engagement in climate change studies and his persuasive communication of the results made an important contribution to policy making. For example, he can convincingly articulate the lessons learned from stratospheric ozone depletion. The ozone hole was a total surprise. No one could have anticipated the catastrophic ozone loss during Antarctic spring. And it cannot be excluded that climate change also holds tipping points where the Earth system swaps from one state to another, perhaps a very undesirable one. These have been central topics since the 1980s, when Paul participated in the German parliamentary commission on “Preventive measures to protect the Earth’s atmosphere.” He helped publish an influential report with compelling arguments that shaped national and international policies on the atmospheric environment and climate change.

Because of his concern about climate change, Paul advocated studies on “geo-engineering”, for example solar radiation management, to investigate if the increase in atmospheric reflectivity could help cool the planet and moderate climate change—just in case impacts might become calamitous. One method is to release sulfur dioxide at high altitudes, much like a volcano eruption, after which the gas is converted into sulfate particles that reflect sunlight and linger in the stratosphere for a few years. The proposal has given rise to controversial discussion. Opponents argue that developing geoengineering options might distract from the real problem, namely reducing greenhouse gas emissions. It should be mentioned that we are still far from practicable geoengineering solutions, and I doubt if one will ever be found, but it cannot hurt thinking about it.

Paul’s comprehensive work on many global change issues has almost inevitably led to the next level of reasoning, as he defined a name for the geological epoch in which it all takes place: the “Anthropocene”. While geologists have traditionally named the most recent 12,000 years the Holocene, Paul argues that in the past centuries the impact of humanity on the Earth’s surface is so large, and unique, that a renaming of the geological timescale is justified. It sends the message that humans have so strongly transformed the planetary environment that it will leave an ineradicable imprint in the rock strata, which will be detectible by future geologists, even if the world population could instantly stop global change. While the proposal on the Anthropocene is still debated among geologists, I conclude that Paul has left an ineradicable imprint in science that will be detectable for a very long time.

Mainz
September 2015

Prof. Dr. Jos Lelieveld
Director, Max Planck Institute for Chemistry
Otto Hahn Institute

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Paul J. Crutzen

Prof. Dr. Paul J. Crutzen during the symposium “The Anthropocene” on 2 December 2013 in Mainz on the occasion of his 80th birthday. Photo by Carsten Costard

Prof. Dr. Paul J. Crutzen thanking the speakers during the symposium “The Anthropocene” on 2 December 2013 on the occasion of his 80th birthday in Mainz. Photo by Carsten Costard

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Part I
On Paul J. Crutzen

Chapter 1
The Background of an Ozone Researcher: A Brief Autobiography

Paul J. Crutzen

1.1 How I Became a Scientist: A Personal History

I was born in Amsterdam on December 3, 1933, the son of Anna Gurk and Jozef Crutzen.[1] I had one sister. My mother's parents moved to the industrial Ruhr region in Germany from East Prussia towards the end of the 19th century. They were of mixed German and Polish origin. In 1929 at the age of 17, my mother moved to Amsterdam to work as a housekeeper. There she met my father. He came from Vaals, a little town in the south-eastern corner of the Netherlands, bordering Belgium and Germany and very close to the historical city of Aachen. He had relatives in the Netherlands, Germany, and Belgium. Thus, from both parents I inherited a cosmopolitan view of the world. Despite having worked in several countries outside the Netherlands since 1958, I have remained a Dutch citizen.

In May 1940 the Netherlands were overrun by the German army. In September of the same year I entered elementary school, "de grote School" (the big school), as it was popularly called. My six years of elementary school largely overlapped with the Second World War. Our school class had to move several times to different premises in Amsterdam after the German army had confiscated our original school building. The last months of the war, between the fall of 1944 and Liberation Day

[1]This text is based on: P.J. Crutzen: "My Life with O_3, NO_x, and Other YZO_x Compounds (Nobel Lecture)", in: *Angew. Chem. Int. Ed Engl.*, **35**, 1758–1777, 1996 and was updated by the author in 2015 Permission to reproduce the original text was granted by the Nobel Foundation in Stockholm in September 2015.

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Paul J. Crutzen as a baby in 1933 in Amsterdam

With his grandmother Elisabeth Gurk in Gelsenkirchen-Buer, 1933 on May 5, 1945, were horrible. During the cold “hongerwinter” (winter of famine) of 1944–1945 there was a severe lack of food and heating fuels. Also water for drinking, cooking, and washing was available only in limited quantities for a few hours per day, which caused poor hygienic conditions. Many died of hunger and disease, including several of my schoolmates. Some relief came at the beginning of 1945 when the Swedish Red Cross dropped food supplies by parachute from airplanes. To welcome them we waved our red, white, and blue Dutch flags in the streets.

Paul J. Crutzen as a pupil in the elementary school in Amsterdam in 1940. *Source* Personal photo collection

In 1945 after a successful exam, I entered the “Hogere Burgerschool” (HBS), Higher Citizen School, a five-year long middle school which prepared students for University. I finished this school in June 1951, with natural sciences as my focal subjects. However, besides Dutch, we all also had to become proficient in 3 foreign languages: French, English, and German. I was given considerable help in learning languages from my parents: German from my mother, French from my father.

Paul J. Crutzen as a boy playing soccer in Amsterdam in 1942. *Source* Personal photo collection

During those years, chemistry definitely was not one of my favorite subjects. They were mathematics and physics, but I also did very well in the three foreign languages. During my school years I spent considerable time at sport: football, cycling, and my greatest passion, skating on the Dutch canals and lakes. I also played chess. I read widely about travels in distant lands, about astronomy, as well as about bridge and tunnel building. Unfortunately, because of a heavy fever, my grades in the final exam of the HBS were not good enough to qualify for a university study stipend, which was very hard to obtain at that time, only six years after the end of the Second World War and a few years after the end of the colonial war in Indonesia, which was a large drain on Dutch resources. As I did not want to be a further financial burden on my parents for another four years or more (my father, a waiter, was often unemployed; my mother worked in the kitchen of a hospital), I chose to attend the "Middelbare Technische School" (MTS), now called the Higher Technical School (HTS), to train as a civil engineer. Although the MTS took three years, the second year was a practical year during which I earned a modest salary, enough to live on for about two years.

From the summer of 1954 until February 1958, with a 21-month interruption for compulsory military service, I worked at the Bridge Construction Bureau of the City of Amsterdam. In the meanwhile, on a vacation in Switzerland I met a sweet girl, Terttu Soininen, a student of Finnish history and literature at the University of Helsinki. A few years later I was able to entice her to marry me. What a great choice I made! She has been the center of a happy family; without her support, I would never have been able to devote so much of my time to studies and science. After our marriage in February 1958, we settled in Gävle, a little town about 200 km north of Stockholm, where I had found a job in a building construction bureau. In December at that same year our daughter Ilona was born. In March 1964, she got a little sister, Sylvia. Ilona is a registered nurse, now living in Boulder, Colorado. Sylvia is a marketing assistant in Munich, Germany. Terttu and I have now three grandchildren.

All this time I had longed for an academic career. One day at the beginning of 1958, I saw an advertisement in a Swedish newspaper by the Department of Meteorology of Stockholm Högskola (from 1961, Stockholm University) announcing an opening for a computer programmer. Although I had not the slightest experience in such work, I applied for the job and had the great luck to be chosen among many candidates. On July 1, 1959, we moved to Stockholm and I started with my second profession. At that time the Meteorology Institute of Stockholm University (MISU) and the associated International Meteorological Institute (IMI) were at the forefront of meteorological research, and many top researchers worked in Stockholm for extended periods. Only about a year earlier the founder of the institutes, Prof. Gustav Rossby, one of the greatest meteorologists ever, had died suddenly and was succeeded by Dr. Bert Bolin, another famous meteorologist, starting director of the Intergovernmental Panel on Climate Change (IPCC). At that time Stockholm University housed the fastest computers in the world (BESK and its successor FACIT).

With the exception of participation in a field campaign in northern Sweden, led by Georg Witt to measure the properties of noctilucent clouds that appear during summer at about 85 km altitude in the coldest parts of atmosphere, and some programming work related to this, I was mainly involved in various meteorological projects until about 1966, especially helping to build and run some of the first numerical (barotropic) weather prediction models.

The great advantage of being at a university department was that I got the opportunity to follow some of the courses that were offered. By 1963 I could thus fulfil the requirement for the "filosofie kandidat" (corresponding to a Master of Science degree), combining the subjects mathematics, mathematical statistics, and meteorology. Unfortunately, I could include neither physics nor chemistry in my formal education, because this would have required my participation in time consuming laboratory courses.

However, around 1965 I was given the task of helping a scientist from the United States to develop a numerical model of the oxygen allotrope distribution in the stratosphere, mesosphere, and lower thermosphere. This project got me highly interested in the photochemistry of atmospheric ozone, and I started an intensive study of the scientific literature. This convinced me of the limited status of scientific knowledge on stratospheric chemistry, thus setting the "initial conditions" for my scientific career. Instead of the initially proposed Ph.D. research project, I preferred