Paul Josef Crutzen (1933–2021)

Scientists across the globe deeply mourn the loss of Professor Paul J Crutzen. He was among the greatest scientific geniuses and pioneers the world has seen in the past century. He passed away on 28 January 2021 in Mainz, Germany and is survived by his wife Mrs Terttu Crutzen, daughters Ilona and Sylvia, and three grandchildren.

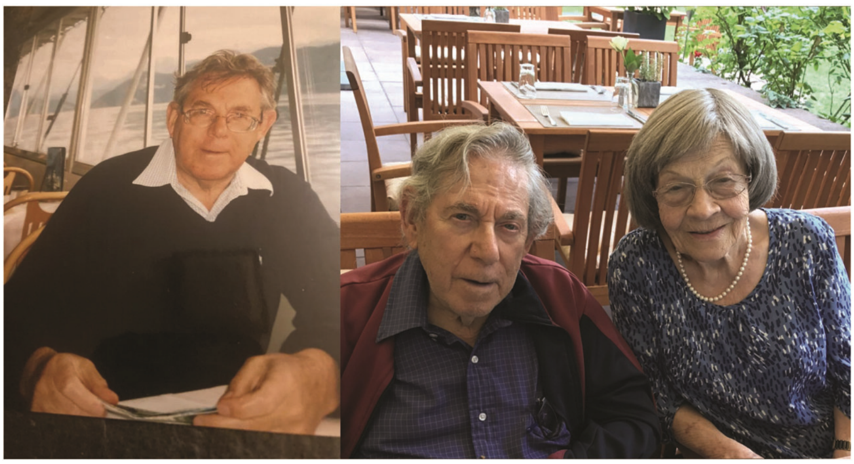
Be it his 1995 Nobel Prize winning work on atmospheric chemistry, which provided the foundation for understanding the natural ozone layer and human-induced damage to it, the habitability for humans on Earth in the aftermath of a nuclear war, perturbations to natural biogeochemical cycles through biomass burning and climate change, climate geoengineering, to name a few major themes[1], he exemplified through his living example that the best scientific attributes (creativity, rigour, critical thinking, sharing of ideas with peers freely) when combined with the best human attributes (compassion, humility, and concern for nature and fellow human beings) could time and again manifest spectacular achievements for the benefit of the world.

Paul's earthly journey commenced on 3 December 1933 in Amsterdam where he was born to Anna Gurk and Jozef Crutzen. While his mother had German and Polish ancestry, his father was Dutch. They belonged to a working-class family and while growing up, conditions were often tough. The brutal winter famine of the 1944–45 and the Second World War were some major hardships that they endured. Paul was an avid sports lover. Playing football, cycling and long-distance skating on lakes were among his favourite activities. He also enjoyed chess, reading about adventurous expeditions and bridges. Due to the cosmopolitan atmosphere at home, he easily became fluent in French, German and English. He liked mathematics and physics too, but chemistry was not among his favourite subjects at school. As he neared completion of school education, he desired to pursue university studies in the natural sciences.

However, in 1951 while appearing for the final school exams, he fell ill and had to take the written and oral exams under high bouts of fever. This affected his final grades rendering him ineligible for pursuing university education with a stipend. As he did not wish to burden his parents financially anymore, he opted for a 3-year Middle Technical School degree in civil engineering. The second year consisted of practical work and enabled him to earn money.

From 1954 to 1959, he worked at the Bridge Construction Bureau of the City of Amsterdam. In 1954 at the age of 21, while vacationing on Mount Pilatus in Switzerland, he met Terttu Soininen from Finland who soon became the love of his life. They fell in love and got married in 1958. In his Nobel lecture[2], Paul made special mention of it as one of the best decisions of his life. He gratefully acknowledged that without Terttu's loving care and extraordinary support, it would have been impossible for him to devote as much time as he did to science. With his characteristic humour, Paul later also remarked that they chose to settle down in Gavle, Sweden after marriage because Sweden happened to be mid-way between Finland and the Netherlands!

Still longing for an academic career, in 1958 when he saw a job advertisement for a computer programmer he promptly applied. To his pleasant surprise, he was selected from amongst many applicants despite no prior experience. On 1 July 1959, he started the new position in the Department of Meteorology at the University of Stockholm. The Head of the Institute was Bert Bolin, who later also served as the first Chairperson of the Intergovernmental Panel on Climate Change. At that time the University of Stockholm housed the fastest computers in the world and for the next five years, Paul helped build and run some of the first numerical weather prediction models, as well as a model for tropical cyclones. Speaking of those times, Paul later reminisced that all code had to be written in machine language and to ensure that computations did not yield wrong results, all operations had to yield numbers in the range of –1 to +1, which furthermore meant that the equations had to be scaled to stay within these limits! While working he started attending theory courses in mathematics, statistics and meteorology and by 1963, he was able to fulfil the requirements for the award of 'Filosofie Kandidat' equivalent to an M.Sc. In 1965, he worked on a project to develop a numerical model for the allotropic distribution of oxygen in the stratosphere, mesosphere and lower thermosphere. This project kindled in him a keen interest to know more about the photochemistry of atmospheric ozone and he embarked on an intensive study of the literature on this topic. Subsequently, for his Ph.D. thesis research ('Filosofie Licentiat'), therefore instead of choosing to further develop the numerical model for tropical cyclones, he chose to work on stratospheric ozone chemistry. The proposal was generously accepted by his Ph.D. Advisor Bert Bolin. In 1968 his thesis work entitled 'Determination of parameters



Left: Paul Crutzen in 2014 during the trip to revisit the place where he had first met Terttu 50 years ago in Switzerland. Right: Paul Crutzen with his wife Terttu when they invited the author's family in Mainz 2017. Photo credit: Vinayak Sinha.

appearing in the "dry" and the "wet" photochemical theories for ozone in the stratosphere' was awarded the highest distinction possible. This work also sowed the seeds for his brilliant paradigm shifting ground breaking subsequent work carried out during his D.Sc. ('Filosofie Docktor') research at Oxford University. Ironically, Paul's motivation to work on stratospheric chemistry was also prompted by a desire to investigate the pure science of 'natural' processes as many of his colleagues were investigating the chemistry of acid rain and damage to ecosystems due to air pollution. What could be more remote and far away from human influence than the stratosphere and what atmospheric region by virtue of its chemical composition and radiation regime could have fewer chemical reactions to worry about – or so he thought!

The British scientist Sydney Chapman had proposed a mechanism[3] in 1930 to explain the photochemical production and loss of ozone by natural processes in the stratosphere through a series of four simple reactions involving only molecular oxygen, oxygen atoms and ozone. Due to the limitations in measuring rates of these reactions accurately and paucity of ozone measurements in the stratosphere, it was thought until the early 1960s that this mechanism was adequate to explain the observed stratospheric ozone abundance. In fact, the shape of the vertical profile of ozone with a maximum in the middle stratosphere was also well predicted by this mechanism. In the 1950s, Bates and Nicolet, together with Chapman pointed out the presence of hydroxyl and hydroperoxyl radicals (collectively called HO_x) in the mesosphere and thermosphere. By the mid 1960s, it was becoming apparent through additional observations and studies that the Chapman mechanism tended to over-estimate the actual abundance of stratospheric ozone significantly. Following up on the HO_x study and laboratory experiments conducted by the 1967 Chemistry Nobel Laureate Norrish, Hampson proposed that HO_x radicals could consume ozone in the upper atmosphere. These were subsequently incorporated into an atmospheric chemical model by Hunt[4]. However, the values they chose for the reaction rate constants were too high. In his Ph.D. thesis work, Crutzen had shown that the choice of the rate constants employed by Hampson and Hunt could not explain the vertical distribution of ozone in the stratosphere and furthermore would result in unrealistically rapid ozone loss in the troposphere. Discarding the theory proposed by Hunt and Hampson to close the stratospheric ozone budget, he noted that the possibility of other photochemical processes should be examined and in particular the influence of nitrogen compounds should be investigated. However, no experimental data existed to test Paul's hypotheses.

After completing his Ph.D. from Stockholm university in 1968 at the age of 35, in 1969 Paul joined as a postdoctoral fellow of the European Space Research Organization in the Clarendon Laboratory of the Department of Physics at the University of Oxford, UK. Aware of Paul's idea concerning the potential influence of nitrogen compounds on stratospheric ozone chemistry, one day the Head of the Research Group John Houghton handed him a solar spectrum taken from a balloon flight by researchers at the University of Denver. This spectrogram indicated the presence of nitric acid in the stratosphere. By analysing it, Paul was able to deduce the approximate amounts and potential distribution of stratospheric nitric acid. But before he could write up the results, Rhine *et al.*[5] published their results on the distribution of nitric acid above 19 km. For nitric acid to occur in the stratosphere, Paul surmised that nitrogen oxides (NO and NO_2) must also be present. Garnering sufficient confidence from these results, in 1970 he proposed that significant ozone loss could occur due to catalytic reactions involving ultra-trace amounts of NO_x in the stratosphere[6]. The source of NO_x was postulated to be photochemical reactions involving nitrous oxide (N_2O), a chemically inert gas produced naturally through microbe mediated nitrification and denitrification processes in soil and water. As it applied later also for anthropogenically emitted chlorofluorocarbons (CFCs), nitrous oxide does not undergo photochemical loss in the troposphere, resulting in lifetimes long enough to be transported to the stratosphere. These ideas proved to be path breaking for the time for more than one reason. The chemistry of nitrogen oxides in the stratosphere had not been considered previously. Also, the idea that a substance present at more than 1000 times lower abundance than stratospheric ozone could be a significant sink for it, appeared somewhat fanciful at first sight. This work also suggested for the first time that the biosphere could affect even the remote stratosphere chemically. This connection brought together biologists and atmospheric scientists and germinated the seed for later unravelling also other biosphere–stratospheric interactions such as those due to carbonyl sulphide and methane. Subsequently, by incorporating more accurate reaction rate constants for reactions in the Chapman mechanism, and the more detailed HO_x and NO_x chemistry, scientists could accurately account for the observed abundance and distribution of stratospheric ozone. Also in 1970, Paul heard about the plans by many countries to deploy large stratospheric fleets of supersonic passenger aircraft through an MIT sponsored study report. The report enabled him to make estimates of how much NO_x might be released into the stratosphere and the resultant ozone loss. The numbers disturbed him greatly, as he could foresee a global environmental problem looming on the horizon because of ozone depletion in the stratosphere. Shockingly, the authors of the report had completely ignored Paul's recent work and concluded in their report that there would be little impact on the stratospheric heat and ozone budgets from the aircraft exhaust emissions. Paul realized that his research area had acquired new importance with major global implications. This motivated him to extend his initial work and publish an updated study[7], incorporating more detailed chemistry of nitrogen oxides. The challenges before him in doing so were immense. For one, unlike today no critically evaluated recommendations for reaction rate constants were available. He pretty much had to compile and comprehend the chemistry on his own which was a major handicap, considering his lack of formal training in chemistry. He was assisted in these efforts by Richard Wayne, a former student of Norrish and fellow co-worker at the University of Oxford.

In 1971 in the US, Harold Johnston of the University of California Berkeley, a physical chemist who measured the rate constants for several reactions relevant to NO_x chemistry published his work in *Science*[8] which corroborated Paul's concerns of potential damage to the natural ozone layer in the stratosphere by a large fleet of supersonic air craft. Harold Johnston encountered fierce criticism from many quarters for this work. The scientific evidence withstood the scrutiny and was irrefutable. Eventually, on account of economic reasons, plans for a large supersonic civilian fleet thankfully got grounded! The work of Crutzen and Johnston led to funding of several new research programmes such as the Climate Impact Assessment Program. In May 1973, Paul submitted his D.Sc. dissertation entitled, 'On the photochemistry of ozone in the stratosphere and troposphere and pollution of the stratosphere by high-flying aircraft' which was again awarded the highest distinction possible.

Then in 1974, Mario Molina and Sherwood Rowland published a paper in *Nature*[9] pointing out that human emitted chemicals called chlorofluorocarbons (CFCs) would undergo photodissociation at short wavelength UV radiation available in the stratosphere and release reactive chlorine atoms that could catalytically destroy stratospheric ozone. In 1985, Farman *et al.*[10] reported observations which showed the formation of a large ozone hole over the Antarctic in springtime. This stunned everyone as the remote and cold and dry Antarctic was literally the last place anyone expected anthropogenic emissions to deplete the ozone layer. Subsequent research by a number of groups including those of Crutzen, Molina and Rowland provided further insights into the mechanism behind the multiphase chemistry. It was shown that the widespread ozone depletion events in the Antarctic in springtime required polar stratospheric clouds (PSCs). PSCs are hydrated nitric acid crystals that form at higher temperatures (~197 K), than normal clouds in the cold and dry wintertime conditions of the isolated Antarctic vortex. Once formed, their surfaces catalyse conversion of hydrogen chloride and chlorine nitrate to chlorine gas (Cl_2) and nitric acid throughout the winter. Then in springtime, when solar radiation becomes available again, photolysis of chlorine releases reactive chlorine atoms that trigger the ozone depletion events. The work of Crutzen, Molina and Rowland provided the scientific basis for the Montreal Protocol, which was signed in 1987. The treaty led to a ban on emissions of CFCs into the atmosphere. As of 2021, it has been ratified by all 197 member countries of the United Nations, making it the most successful global treaty.

From 1974 to 1980, Paul Crutzen worked at the National Centre of Atmospheric Research (NCAR) and National Oceanic and Atmospheric Administration (NOAA) in Boulder, Colorado, USA. By 1980, he had become the Director of the Air Quality Division at NCAR. Also as an Adjunct Professor at the Colorado State University, he actively participated in the teaching activities. During this period, motivated by Hiriam Levy's work on tropospheric hydroxyl radicals[11], together with his then Ph.D. students Susan Solomon and Jack Fishman he started focussing on tropospheric chemistry. A number of new findings[12–14] resulted from these efforts which completely changed the then understanding of tropospheric ozone and also pointed to its role as a potent green-house gas. Understanding of tropospheric ozone and its chemical production through organic trace gases and nitrogen oxides, helped clear lingering notions about downward transport of ozone from the stratosphere being the major source of tropospheric ozone. While nitrogen oxides acted as a sink of ozone in the stratosphere, in the presence of reactive organic trace gases, they acted as a source of ozone in the troposphere.

In 1980, Paul Crutzen returned to Europe to succeed Christian Junge as the new Director of the Atmospheric Chemistry Department at the Max Planck Institute for Chemistry in Mainz, Germany. In 1981 he was asked by the editor of *Ambio* to contribute to a special issue on the environmental consequences of a major nuclear war. Together with John Birks, he carried out research on the politically sensitive and surcharged issue. They pointed out that an atomic war would cause huge fires to run through forests and cities, and the sooty smoke particles from the fires would absorb sunlight, leading to the surface of the earth becoming dark and cold[15]. There would be widespread starvation. Immediate deaths due to the atomic bomb would be soon exceeded by deaths due to the indirect climatic consequences, even in regions that did not go to war. Paul considered this idea to be amongst the most important he ever had in his life[16].

From 1980 to 2000, he set up an active international research programme at the Max Planck Institute of Chemistry. He conducted numerous atmospheric field measurement studies in diverse ecosystems of the world and used the data in advanced numerical models. Many of these studies provided the first findings/discoveries and serve as the basis for our present day understanding of atmospheric chemistry processes, sources and sinks of green-house gases and volatile organic compounds. In 1995, Paul Crutzen was jointly awarded the Nobel Prize in Chemistry with Mario Molina and Sherwood Rowland for their work on atmospheric chemistry, with special mention of their work concerning the formation and decomposition of ozone.

Perhaps also worth mentioning here is the multi-nation Indian Ocean Experiment conducted in 1999 in which Paul's division partnered with Veerabhadran Ramanathan of Scripps Institution of Oceanography[17]. This international effort also involved leading Indian scientists like A. P. Mitra of National Physical Laboratory, D. R. Sikka of Indian Institute of Tropical Meteorology, Pune and Shyam Lal of the Physical Research Laboratory, Ahmedabad. A large number of young Indian scientists, who are leading lights of Indian atmospheric science today such as S. K. Satheesh of the Indian Institute of Science, Bengaluru also benefitted from participation in INDOEX. The findings triggered several new research programmes and national studies spearheaded by ISRO under K. Krishnamoorthy and Shyam Lal. These have contributed immensely to improve the current understanding of the nexus between air pollutant emissions and climate over the Indian region.

The term Anthropocene now used in multiple disciplines is credited to Crutzen. The story[18] goes that at a Scientific Committee Meeting of the International Geosphere–Biosphere Program (IGBP) in Mexico in 2000, while listening to the review of the unparalleled global environmental change that had occurred in recent decades of the Holocene (a geological epoch which started ~12,000

Paul Crutzen with his grand-daughter Lea in 2004. Photo credit: Mrs Terttu Crutzen.

years ago after the last Ice Age), Paul felt exasperated and he suddenly remarked aloud: 'We are no longer in the Holocene but in (with pause) – the Anthropocene.' The spontaneous improvisation caught the attention of the audience, crystallizing the growing realization that the Earth system had recently begun to change at a much more dramatic rate and scale than through many previous millennia of slowly growing human occupation of the planet. Crutzen invited Stoermer, an ecologist who had first used the term in an ecological context to co-publish the term and concept in the newsletter of the International Geosphere and Biosphere Program[19]. Two years later he published the article entitled Geology of mankind – The Anthropocene in Nature[20]. The concept of a planet that is human-dominated on a *geological* scale had been suggested previously too, but never considered seriously by mainstream geologists. For his 80th birthday celebrations in 2013 in Mainz, at the scientific symposium organized in his honour, he chose Anthropocene as the theme. When the Anthropocene Working Group members met him in Mainz in 2018, even though he had not been keeping well for many weeks, he still engaged in two long days of detailed evidence and disputation and in his characteristic fashion did not show the least attachment to his brainchild. As stated by one of the members of the group, 'He was unfailingly supportive and encouraging about the work being done, even when the procedural nitpicking of geological timescale work went against the grain of his own quickness and clarity. Not all great scientists are likable and good company – Crutzen was.'

In 2006, he wrote a provocative editorial on climate geo-engineering[21], which appeared to be rather controversial. Speaking about his motivation in this regard in an interview[16] he noted: 'I wanted to give people a wake-up call. I wanted to make it clear what serious compromises we are making to the climate and to atmospheric chemistry. If we carry on this way the consequences cannot be overseen. The most important thing is that we reduce CO_2 emissions. That is the number one issue. But we haven't seen anything of it yet. And that is why I wanted to make it quite clear how dangerous the situation is.'

During his illustrious career, Paul J. Crutzen published over 360 peer-reviewed scientific journal articles, 135 scientific publications in research journals, and 15 books. He was one of the most highly cited scientists in the world, having been bestowed numerous awards and honours, as a member of many scientific academies. He has a street in Holland and comet named after him. He was a recipient of the Jawaharlal Nehru Birth Centenary Medal 2006, awarded by the Indian National Science Academy, New Delhi, India. In addition to his scientific body of work, another lasting legacy are the large number (46) of Ph.D. students and Postdocs who trained directly under him, many of whom like Jos Lelieveld (who succeeded him as Director of Max Planck Institute for Chemistry in Mainz, Germany), Frank Dentener (at European Commission's Joint Research Centre, Italy), Mark Lawrence (Director at Institute for Advanced Sustainability Studies, Potsdam, Germany), John Burrows (Director of the Institutes of Environmental Physics and Remote Sensing at the University of Bremen) and Susan Solomon (Lee and Geraldine Martin Professor of Environmental Studies at MIT, USA) have gone on to become famous leaders of the field and continue to reshape and redefine research pertaining to atmospheric chemistry, global environmental change, climate of the earth and air pollution.

Crutzen cared for and continued to actively support outreach and training of early career scientists across the world well after his retirement through training schools, special talks and events including for school children. There are hundreds of students spanning several generations in developing nations of Asia, Africa and South America whose own research careers have been touched and benefitted by Paul Crutzen's warm and generous scientific feedback and inspirational mentoring. I still recollect how in 2006, while presenting measurements on methane emissions from forested ecosystems during the departmental meeting held at Schloss Ringberg, Paul got as excited as the Ph.D. student author about the ambient methane data collected from the boreal forest in Finland and tropical rain forest in Suriname. Earlier that year, Frank Keppler had reported the surprising finding in *Nature*[22], that terrestrial plants could emit methane under aerobic conditions. The experience of working with Paul is deeply treasured by me as it gave a first-hand experience of the meticulous and friendly manner in which he contributed to his co-authored scientific papers. When I got back the draft of the manuscript from Paul, I found that Paul had checked each and every reference manually by striking out the citation in the reference list whenever he first read it in the main text. The manuscript was published as a preprint[23] and received critical scrutiny. The most valuable lesson however was while preparing the revised submission, I realized that a previously unrecognized temperature effect in the inlet could explain most of the observed night-time methane variability, which had been used to derive the flux. I was rather crestfallen as it was only my second first author paper, but Paul put me completely at ease by saying, 'Vinayak I should have also thought about it before we submitted the paper but it is good that we now know about how strong this effect can be so we can be more careful in the future.' My Ph.D. advisor and the other senior co-author were also amazingly supportive. The lesson learnt was that it is ok to make 'honest' mistakes and the process of science allows for acknowledging past errors and moving on after being the wiser for it.

Paul dedicated his Nobel lecture to the generation of his grandchildren and future generations hoping that they would get to celebrate the recovery of the Antarctic ozone hole. He also tended to end his lectures with a picture of one of his grandchildren and himself to remind people of their duty to hand over a safe planet to subsequent generations.

I would like to end this tribute to him by quoting from one of the condolences[24] posted in memory of Paul J. Crutzen: 'It is very sad that Paul has died but we all have reason to be thankful that he lived.'

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