Paul Jozef Crutzen
3 December 1933 – 28 January 2021
Elected ForMemRS 2006

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Abstract

Paul J. Crutzen was both a warmhearted person and a brilliant scientist. His research interests were broad, encompassing topics of relevance in the mesosphere, the stratosphere and the troposphere. He made fundamental scientific contributions to a wide range of topics in the science of all these atmospheric regions. For example, he first described the NO_x driven ozone loss cycle in the stratosphere, he contributed key ideas on how to explain the "ozone hole" and he made fundamental discoveries on the impact of biomass-burning on the atmosphere. Understanding and combating the origins of air pollution and climate change were driving motivations for his life's work. Further, he pioneered the concept that is known as "nuclear winter", he initiated the resumption of discussion on geoengineering and coined the term "Anthropocene". With all this focus on science, he would not forget the importance of private life, both as regards his family and his colleagues and students. In 1995, together with Mario J. Molina, and F. Sherwood Rowland, Paul was awarded the Nobel Prize in Chemistry for ground-breaking work "in atmospheric chemistry, particularly concerning the formation and decomposition of ozone". Finally, Paul's work not only had a profound influence on the scientific world, but also on different aspects of environmental politics throughout many countries. His works on the impact of human activities on the atmosphere and climate have been influential in the past and are influential today. They provide a beacon of knowledge for the next generations of scientists and environmental policymakers.

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A portrait of Paul Crutzen

Figure 1: Portrait of Paul Crutzen for the Royal Society. (Picture courtesy Royal Society.)

1 Early life and education

Figure 2: Paul Crutzen with his father. (Picture courtesy of Ilona Crutzen.)

Paul Jozef Crutzen was born in Amsterdam on 3 December 1933; he was the son of Anna Gurk and Jozef Crutzen (Fig. 2). His younger sister Elisabeth Helena (Lies) Crutzen was born on 29 February 1936; she lived all her life in Amsterdam. She passed away in 2003. His mother's parents were of mixed German and Polish origin and lived in the Ruhr region in Germany. His father had relatives in the Netherlands, Germany, and Belgium. Through his family and relatives, Paul inherited a cosmopolitan view of the world at an early age.

Paul had a hard youth; his six years of elementary school largely overlapped with the Second World War. Particularly horrible was the cold winter 1944-1945 known as "hongerwinter" (winter of famine). In this winter many Dutch died of hunger and disease, including several of Paul's schoolmates (reference 30) [1].

In 1946 Paul entered the "Hogere Burgerschool" (Higher Citizen School), which he finished in June 1951. His academic focus lay in the natural sciences, but he also had to become proficient in French, English, and German. During these years at school, he

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[1] Numbers in this form refer to the bibliography at the end of the text.

spent considerable time doing sport: football, cycling, and – his greatest passion – long distance skating on the Dutch canals and lakes. All through his life, he kept a close eye on the success of the Dutch skating team at international sports events.

Because of a heavy fever, his grades in the final exam of the Higher Citizen School were not sufficient to qualify him for a university stipend. But as he did not want to continue to be a financial burden on his parents, he chose to attend the “Middelbare Technische School” (middle technical school) to train as a civil engineer, which allowed him to earn a modest salary in the second year of school (30, 45).

Figure 3: Terttu and Paul Crutzen on their wedding day in Amsterdam on 14 February 1958. (Picture courtesy of Ilona Crutzen.)

From the summer of 1954 until February 1958, with a 21-month interruption for compulsory military service, Paul worked at the Bridge Construction Bureau of the City of Amsterdam. During this time, on a vacation trip to Switzerland, he met Terttu Soininen, a student of Finnish history and literature at the University of Helsinki. They married in February 1958 (Fig. 3) and moved to Sweden, to the small town Gävle (about 200 km north of Stockholm) where Paul had found a job in a building construction bureau. In December of that year their daughter Ilona was born and in March 1964, her sister Sylvia. Paul and Terttu now have three grand children.

2 Starting an academic career

2.1 Stockholm

The opportunity for an academic career arose in 1958, when Paul Crutzen saw an advertisement in a Swedish newspaper by the Department of Meteorology of Stockholm Högskola (now Stockholm University) announcing an opening for a computer programmer. He had no experience in such work, but applied for the job and was chosen (30, 45). So on 1 July 1959 Terttu and Paul moved to Stockholm. This was the start of Paul's scientific career.

At the Meteorology Institute of Stockholm University (MISU) he met many eminent scientists; the director of the institute was Bert Bolin, who later became director of the Intergovernmental Panel on Climate Change (IPCC). Bert Bolin and Georg Witt, an expert on the aeronomy of the upper atmosphere, were Paul's supervisors at MISU. Whilst at MISU, he met his fellow student Henning Rodhe, now (retired) Professor in Chemical Meteorology at MISU, who became a close friend for life.

At that time, Stockholm University housed a computer called BESK, which was among the fastest in the world. However, computer programming was very different from what it is today; coding had to be done in machine language and not even the first variants of Fortran were available. But along with this work Paul followed his scientific interest; in 1963 he could thus fulfil the requirements for a “filosofie kandidat” (Master of Science) degree and, around 1965, got the task of helping J. R. Blankenship, a visiting scientist from the US, to develop a numerical model of the distribution of oxygen in different structural forms in the stratosphere, mesosphere, and lower thermosphere. This was the beginning of Paul's research in atmospheric chemistry and also resulted in his first scientific paper (1). Stratospheric ozone also became the topic of his “filosofie licentiat thesis” (PhD thesis). It is interesting to note that at that time at MISU, the issue of human impact on the atmosphere through “acid rain” was a hot topic, but that Paul decided to stay away initially from research addressing human impact on the atmosphere and decided “to do pure science related to natural processes and therefore [. . .] picked stratospheric ozone” (30) – this decision would be revisited later.

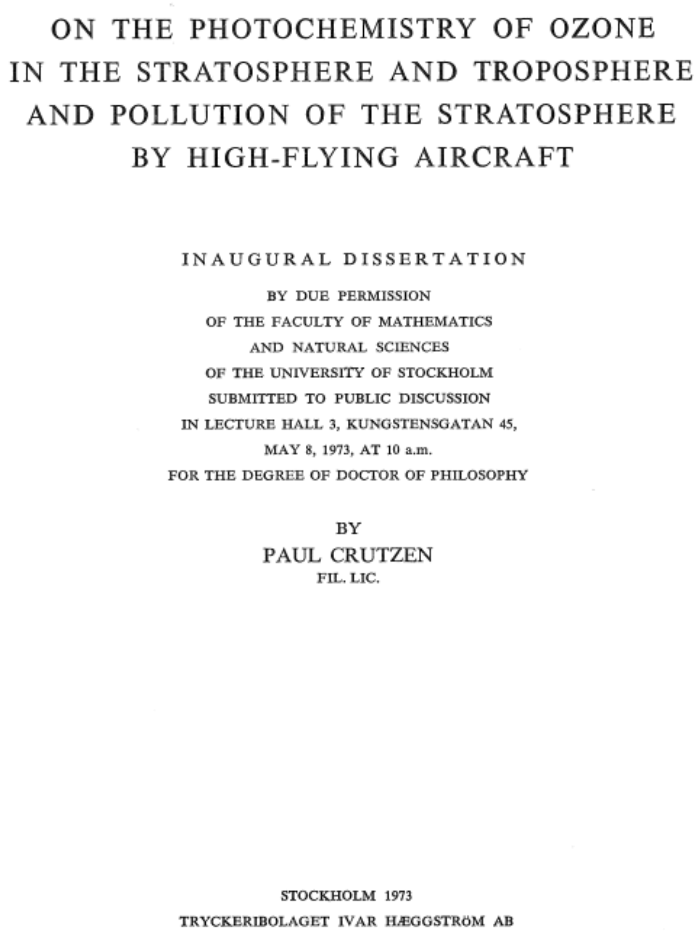
2.2 Filosofie Doctor

In May 1973, Paul Crutzen submitted his inaugural dissertation entitled “On the Photochemistry of Ozone in the Stratosphere and Troposphere and Pollution of the Stratosphere by High-Flying Aircraft” to the Faculty of Natural Sciences of Stockholm University. His thesis was based on four publications (references 3, 4, 5 and 6, with the last paper being in press at the time of submission).

The exam for the “filosofie doctor” took place in lecture hall 3, on 8 May 1973 at ten in the morning. The title page of his thesis is reprinted here (Fig. 4). At that time it was obligatory to have “opponents” of the candidate; in Paul's exam these were Dr. John Houghton (later Sir John) and Dr. Richard Wayne of the University of Oxford, who wore their university gowns on the occasion; Figure 5 shows a picture of Paul together with Dr. John Houghton (wearing an Oxford gown) and Dr. Richard Wayne (wearing a Cambridge gown) after the exam.

3 The chemical balance of the ozone layer

Paul Crutzen was the first to suggest that reactions catalysed by NO and NO_2 control the ozone concentrations in the middle stratosphere (reference 3); in 1970 he put forward the



ON THE PHOTOCHEMISTRY OF OZONE
IN THE STRATOSPHERE AND TROPOSPHERE
AND POLLUTION OF THE STRATOSPHERE
BY HIGH-FLYING AIRCRAFT

INAUGURAL DISSERTATION
BY DUE PERMISSION
OF THE FACULTY OF MATHEMATICS
AND NATURAL SCIENCES
OF THE UNIVERSITY OF STOCKHOLM
SUBMITTED TO PUBLIC DISCUSSION
IN LECTURE HALL 3, KUNGSTENSGATAN 45,
MAY 8, 1973, AT 10 a.m.
FOR THE DEGREE OF DOCTOR OF PHILOSOPHY

BY
PAUL CRUTZEN
FIL. LIC.

STOCKHOLM 1973
TRYCKERIBOLAGET IVAR HÆGGSTRÖM AB

Figure 4: The title page of the inaugural dissertation of Paul Crutzen submitted in May 1973. This was one of the last occasions in which the classical “Filosofie Doktor” (similar to the Habilitation in other countries) was awarded (30).

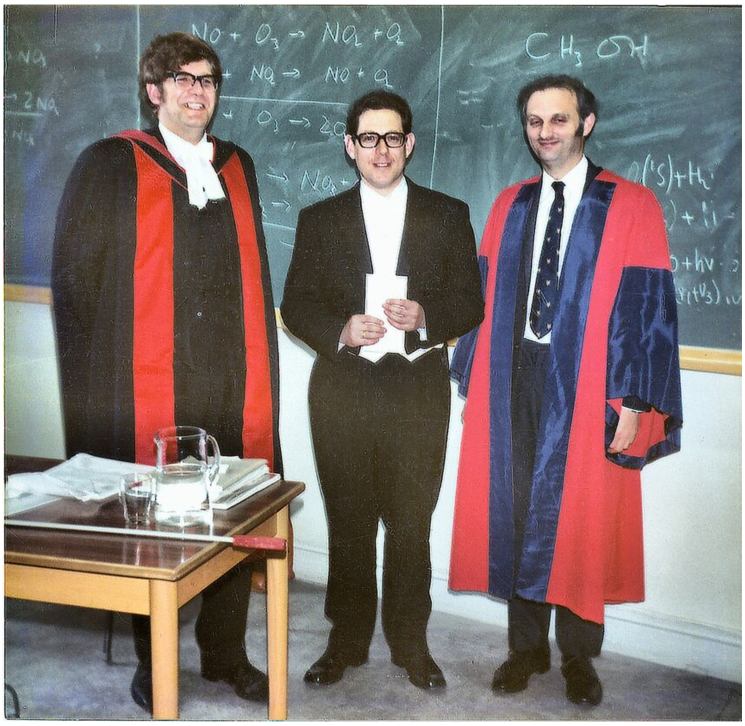


Figure 5: After Paul's exam in Stockholm in 1973. From left to right: Richard P. Wayne, Paul Crutzen, John Houghton (later Sir John). Note reactions (1) and (2) on the blackboard. Picture was taken with the camera of R. P. Wayne, probably by G. Witt. (Picture courtesy of Richard P. Wayne, see also reference 45.)

following catalytic cycle:

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\begin{aligned}
NO + O_3 &\longrightarrow NO_2 + O_2 \qquad (1)\\
NO_2 + O &\longrightarrow NO + O_2 \qquad (2)
\end{aligned}
$$

where the sum of NO and NO_2 is defined as $NO_x = NO + NO_2$ and O indicates an oxygen atom in ground-state ($O(^3P)$).

The discovery of the mechanism was a major achievement and an important step towards facilitating a quantitative description of the chemistry of the stratospheric ozone layer. It formed part of the basis of Paul being awarded the Nobel Prize in Chemistry in 1995. In 1970, many uncertainties regarding stratospheric nitrogen remained; in particular the first measurements of nitrogen compounds in the stratosphere had just become available (Murcray et al., 1968; Rhine et al., 1969). Today it is established that reactions (1) and (2) are the most important ozone loss reactions in the stratosphere in the altitude region between about 25 and 45 km (e.g. Portmann et al., 2012, see also references 29, 30).

In the early 1970s, the tools for numerical simulations were one-dimensional column models for mid-latitude conditions (30°N), with about a 1 km vertical resolution, where the entire atmospheric transport was described by an empirical vertical eddy diffusion coefficient (K_z) (Brasseur, 2019). Research on the chemical balance of the ozone layer had started with the British scientist Sydney Chapman (1930) who proposed that the formation of ozone is initiated by the photolysis of O_2, by solar radiation at short wavelengths

$$O_2 + h\nu \longrightarrow 2\,O \qquad (\lambda \leq 240\,\mathrm{nm}) \qquad (3)$$

followed by the association process

$$O + O_2 + M \longrightarrow O_3 + M \qquad (4)$$

(where M is a collision partner such as atmospheric N_2, O_2 or CO_2 that can remove energy released in the association reaction). The photodissociation of O_3 produces O

$$O_3 + h\nu \longrightarrow O + O_2 \qquad (\lambda \geq 320\,\mathrm{nm}) \qquad (5)$$

but does not constitute a loss of O_3 because there is a steady state between O and O_3 (30). Therefore, “odd oxygen” O_x is defined as $O_x = O + O_3$. Destruction of odd oxygen occurs through the reaction

$$O + O_3 \longrightarrow 2\,O_2 \quad . \qquad (6)$$

Both the production and destruction of an odd oxygen species are equivalent to the production and destruction of an ozone molecule. This scheme, referred to as the “Chapman reactions” was considered for many years to be the theoretical basis for a quantitative description of stratospheric ozone, including its vertical distribution in the stratosphere (see e.g. reference 30). However, this scheme (Chapman, 1930) greatly overestimates ozone concentrations compared to measurements and can not explain the observed latitudinal distribution of ozone (Brasseur, 2019).

Twenty years later, Bates and Nicolet (1950) suggested that reactions involving OH and HO_2 radicals should lead to significant catalytic ozone loss in the mesosphere and thermosphere. Further studies focused on the impact of HO_x-catalysed reactions (HO_x = OH + HO_2 + H) on stratospheric ozone and new measurements of the rate constants for the Chapman reactions were conducted (e.g. Müller, 2009; Brasseur, 2019, and references therein; see also reference 30).

Paul, in his “filosofie licentiat” thesis of 1968 concluded that these earlier theories could not explain the vertical distribution of ozone above 25 km (and in the troposphere) and that other photochemical processes beyond OH and HO_2 radicals should be introduced into the theory of stratospheric ozone (2, 30). Two years later, in 1970, Paul demonstrated that NO_x species (i.e. reactions (1) and (2) are essential for the chemical balance of the stratospheric ozone layer (3).

In summer 1969 Paul had joined the Department of Atmospheric Physics at the Clarendon Laboratory of Oxford University as a postdoctoral fellow. He stayed there for a two year period and returned to Stockholm in July 1971. In Oxford, the head of the research group, Sir John Houghton, supported Paul’s idea about the potential role of NO_x in stratospheric ozone chemistry. At that time the first measurements of nitrogen compounds in the atmosphere and their implications for the source of nitrogen in the stratosphere were a focus of research (see e.g. references 4 and 30); we know much more today (sections 4.2 and 4.3).

4 The impact of nitrogen on stratospheric ozone

4.1 Supersonic transport

In a paper on the vertical distributions of trace gases in the O_x/NO_x/HO_x/HNO_x system in 1971 (4) Paul Crutzen discussed the difficulties of doing model calculations in the absence of key information on chemical rate coefficients. But he also stated that the increase of NO_x caused by the planned supersonic transport in the stratosphere should lead to observable changes in stratospheric ozone.

In the early 1970s a controversy about supersonic transport had started in the United States. At that time, large fleets of stratospheric supersonic aircraft were considered (US: Boeing, Britain/France: Concorde, Soviet Union: Tupolev) and a fleet of 500 supersonic planes seemed a reasonable estimate. The concern was an enhanced catalytic ozone destruction. Originally ozone destruction by OH and HO_2 radicals (resulting from the release of water vapour in the engine exhausts) was considered, but soon it was realised that the catalytic destruction of ozone by NO_x posed a much greater threat to the ozone layer (Johnston, 1971).

In 1972, Paul made an important statement on the issue of supersonic transport (5). He made this statement despite (or perhaps because of) the limited knowledge on stratospheric processes at this time. He said that the “minimum requirement is therefore that extensive supersonic air traffic should not take place in the stratosphere before reliable predictions can be made of the possible environmental consequences of such operations”.

4.2 Solar Proton Events

In August 1972, a major solar proton event occurred, during which, within a few hours, large quantities of NO were produced at high latitudes (17). Paul showed, together with I. S. A. Isaksen and G. C. Reid, that solar proton events should lead to a depletion of stratospheric ozone (9). A substantial depletion of ozone was indeed observed after the solar proton event in 1972 (about 20% between 40 and 45 km) and could be reproduced by model calculations by Susan Solomon and Paul (17); these calculations also considered chlorine chemistry. Thus, the analysis of a natural phenomenon provided further support for the impact of catalytic reactions involving NO_x on stratospheric ozone (reactions 1 and 2).

4.3 Nitrogen fertilisers

Paul also established that the biosphere has a direct impact on stratospheric ozone; the biosphere (i.e. the soil) is a source of N_2O and therefore a source of stratospheric NO_x (4, 11). In the early 1970s many aspects of nitrogen chemistry in the stratosphere were unknown or at least uncertain. However, it soon became clear that the reaction

$$N_2O + O(^1D) \longrightarrow 2\,NO \tag{7}$$

constitutes an important source of NO_x in the stratosphere (e.g., McElroy and McConnell, 1971, see also reference 4). The dominant mechanism for loss of N_2O in the stratosphere is photolysis

$$N_2O + h\nu \longrightarrow N_2 + O \tag{8}$$

which, however, does not produce NO_x.

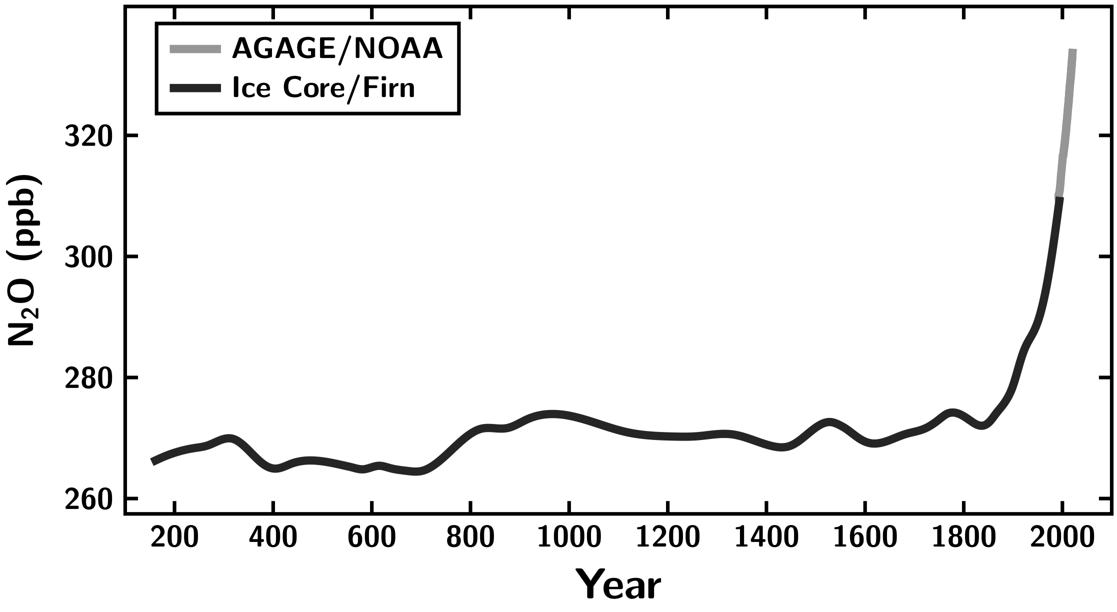

Figure 6: The temporal development of atmospheric concentrations of N_2O over the past 2000 years. (Updated to the year 2021 from Müller, 2021). Historical data (dark line, MacFarling Meure et al., 2006; Park et al., 2012; Rubino et al., 2019) are deduced from ice core and firn measurements; data after 1995 (light grey line) are global mean data from the atmospheric monitoring networks AGAGE (Prinn et al., 2018) and NOAA (Dlugokencky et al., 1994). (See acknowledgements for the data sources).

N_2O is a natural product of microbiological processes in soils and waters; but human activities, such as the application of nitrogen fertilisers in agriculture, cause substantial additional emissions of N_2O to the atmosphere (Park et al., 2012). The point of the impact of human activities on the atmospheric nitrogen budget was emphasised in 1977 in another influential paper by Paul and Dieter Ehhalt (reference 11, see also Solomon, 2021b).

We know today that the most substantial source of stratospheric NO_x (and other nitrogen compounds) is N_2O, which is emitted at the surface of the earth and which is essentially inert in the troposphere. Further, human activities have led to a significant increase in atmospheric concentrations of N_2O (Park et al., 2012). Concentrations of N_2O have risen from levels below 280 ppb[2] before 1800 to more than 334 ppb today (Fig. 6). In the second half of the 21st century, when chlorine containing ozone depleting species will be substantially reduced (see section 6.3), the temporal development of N_2O (together with CO_2 and CH_4) will constitute the dominant driver of changes of stratospheric ozone (Ravishankara et al., 2009; Portmann et al., 2012; Solomon, 2021b).

5 Tropospheric Chemistry

5.1 Tropospheric ozone

The tropospheric ozone budget is influenced both by a downward transport of ozone from the stratosphere to the troposphere and by in situ tropospheric reactions. Paul Crutzen, in 1978/1979, together with Jack Fishman and Susan Solomon (30), presented evidence for large *in situ* photochemical sources and sinks of tropospheric ozone (12, 14). And tropospheric ozone is not only important as a key component of both pristine and polluted air; it is also a major greenhouse gas (16).

Paul contributed significantly to scientific questions regarding tropospheric ozone (e.g., 12, 14, 16, 24). He turned his attention to tropospheric chemistry, a topic which he had hitherto not focused on (30) following the publication of the important paper by Levy (1971). In the troposphere the chain of reactions

$$\begin{aligned} RO_2 + NO &\longrightarrow RO + NO_2 && && (9)\\ NO_2 + h\nu &\longrightarrow NO + O && (\lambda \leq 405\,\mathrm{nm}) && (10)\\ O + O_2 + M &\longrightarrow O_3 + M && && (4) \end{aligned}$$

is the basic mechanism of chemical ozone formation (here $RO_2 = HO_2, CH_3O_2$, or another organoperoxy radical and M is a collision partner, see reaction 4).

Starting with the paper by Levy (1971), the importance of the photolysis of O_3 by solar ultraviolet radiation was realised, which splits O_3 into an O atom and an O_2 molecule. The O atom is formed in an (electronically) excited state, $O(^1D)$, and carries enough energy to allow it to abstract hydrogen from H_2O yielding two OH radicals:

$$O(^1D) + H_2O \longrightarrow 2\,OH \qquad (11)$$

a process energetically unfavourable for ground-state oxygen atoms ($O(^3P)$). $O(^1D)$ can react similarly with other hydrogen-bearing molecules, such as hydrocarbons. Levy (1971) proposed that O_3 photolysis followed by reaction (11) should be a source of OH radicals in the troposphere, which are responsible for the tropospheric oxidation of CH_4

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[2] Parts per billion, a measure of the molar mixing ratio.

and CO. The different roles of CH_4 and CO in tropospheric chemistry and further details of Paul's discoveries in this field are described by Fishman et al. (2022). OH radicals in the troposphere are also responsible for reaction with a variety of other compounds, thus initiating the oxidation of these species and their removal from the atmosphere.

In a model study in 1991, Paul together with Peter H. Zimmermann (24), calculated the OH concentrations for pre-industrial and present conditions; they reported that tropospheric OH reacts rapidly with the majority of gases that are emitted to the atmosphere because of natural processes and human activities. Through the reaction with OH, these gases are removed from the atmosphere. This approach allowed changes in O_3, CO, and OH concentrations to be determined that have occurred as a result of human activity (24). The fact that the highest tropospheric OH concentrations are found in the tropics (30), points to the importance of the tropics in atmospheric chemistry (see also section 5.3).

5.2 Tropospheric pollution

Paul also contributed to research on the impact of HNO_3 and H_2SO_4 to acidic precipitation (18) and initiated major measurement campaigns on tropospheric pollution in Asia and in the Mediterranean (39, 43). The Indian Ocean Experiment found surprisingly high pollution levels over the entire Northern Indian ocean region during the dry season January–March 1999. In this time period, South and Southeast Asian emissions of pollutants resulted in poor air quality over a huge area (39). The Mediterranean Intensive Oxidant Study (MINOS), in the summer of 2001, uncovered air pollution layers in the Mediterranean from the surface to an altitude of 15 kilometres. Pollution was found to be transported to the Mediterranean from European, Asian and (to a lesser extent) North American sources (43).

5.3 Biomass-burning

First estimates by Paul and a number of colleagues in 1979 (15) showed a major impact of a number of trace gases originating from biomass-burning (e.g., CO, H_2, NO, CH_3Cl and COS) on the atmosphere. These estimates were based on measurements at the NCAR laboratories in Boulder (15, 30) and demonstrated that emissions of these gases from biomass-burning could constitute a significant fraction of their total global emissions (15). A year earlier, Pat Zimmerman and colleagues (including Paul) had already pointed out that the release of hydrocarbons (isoprene and terpenes) from biomass burning could contribute in an important way to the atmospheric sources of CO and H_2 (13).

These first results stimulated a considerable international research effort including several measurement campaigns in the tropics (e.g. 39). It was found that large quantities of ozone are formed as a result of biomass-burning emissions (22, 30) and that biomass-burning emissions in South and Southeast Asia strongly enhance tropospheric CO concentrations (39). Moreover, biomass-burning is an important source of (different types of) aerosol particles. Together with Andi Andreae, Paul showed that such particles may serve as cloud condensation nuclei and thus substantially influence cloud microphysical and optical properties (22). This has consequences for the radiation budget and the hydrological cycle of the atmosphere (22).

Today, the importance of biomass-burning for the atmosphere is well established.

Biomass-burning is widespread, especially in the tropics; it is a source of a great number of photochemically and radiatively active trace gases, such as NO_x, CO, CH_4, reactive hydrocarbons, H_2 N_2O, OCS, CH_3Cl, and HCN. Biomass-burning is not restricted to tropical deforestation, but is also a common activity related to agriculture (particularly in developing countries) and fuel wood combustion; it may also happen through smouldering fires in peatlands.

6 Anthropogenic chlorine in the stratosphere and the Antarctic ozone hole

6.1 Global ozone loss

In 1973, Paul Crutzen received the preprint of a paper entitled "Stratospheric sink for chlorofluoromethanes: chlorine atom-catalysed destruction of ozone" by M. J. Molina and F. S. Rowland, neither of whom he had heard of before (30, 53). Their paper (Molina and Rowland, 1974) pointed out that the chain of reactions

$$\mathrm{Cl} + \mathrm{O_3} \longrightarrow \mathrm{ClO} + \mathrm{O_2} \qquad (12)$$

$$\mathrm{ClO} + \mathrm{O} \longrightarrow \mathrm{Cl} + \mathrm{O_2} \qquad (13)$$

could destroy ozone in the stratosphere. The stratospheric chlorine, in turn, would originate from the photolysis of industrially produced chlorofluorocarbons (CFCs) at stratospheric altitudes. Further, their paper made the point that the reduction in stratospheric ozone would result in an increase in the amount of UV reaching the earth's surface, with potential severe environmental consequences (Harris, 2020).

Paul immediately understood the importance of this paper and about two months after the publication by Molina and Rowland (1974) presented a model analysis (7) on the potential ozone depletion resulting from continued use of CFCs; the paper indicated the possibility of up to $\sim 40\%$ ozone depletion near 40 km altitude as a result of continued use of CFCs at 1974 rates.

In the following years the paper by Molina and Rowland (1974) stimulated the making of a great deal of new measurements in the laboratory and in the atmosphere as well as major advances in atmospheric modelling (Harris, 2020). Meanwhile, in 1974, Paul had moved with his family to Boulder, Colorado, where he assumed two part-time positions; one as a consultant at the Aeronomy Laboratory of the National Oceanic and Atmospheric Administration (NOAA) and one at the Upper Atmosphere Project of the National Center for Atmospheric Research (NCAR). Later, in 1977, he took up the directorship of the Air Quality Division of NCAR. In 1980 he moved back to Europe to take up a directorship at the "Max-Planck-Institut (MPI) für Chemie" in Mainz as the successor of Christian Junge. Figure 7 shows him at his desk in his office at the "MPI für Chemie". In Mainz you could see him walk to work with his little rucksack in the morning; he did not own a car.

6.2 Polar ozone loss

In the early 1980s, Paul was heavily involved in various studies on the potential environmental impacts of a major nuclear war (see section 7). But in 1985 came another

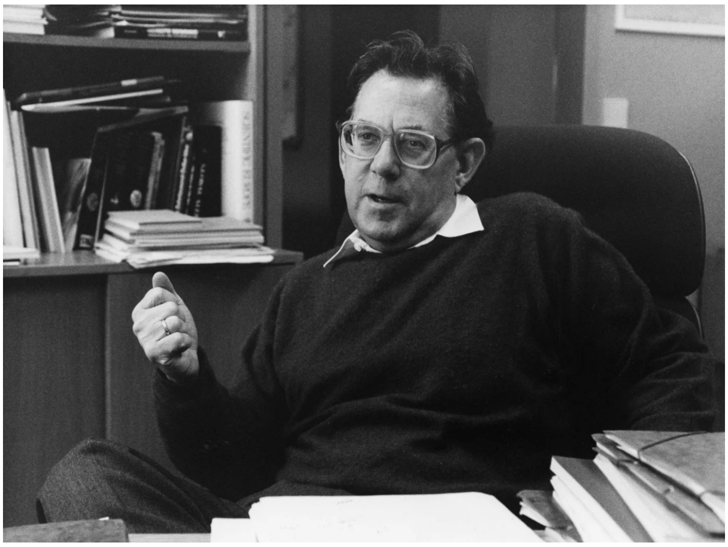

Figure 7: Paul Crutzen in his office at the “Max-Planck-Institut (MPI) für Chemie” in Mainz in 1994. (From the archives of the “Max-Planck-Gesellschaft”, Berlin-Dahlem).

major (and surprising) discovery – J. Farman, B. Gardiner, and J. Shanklin of the British Antarctic Survey reported that the spring values of total ozone in Antarctica since the late 1970s had fallen considerably (Farman et al., 1985); this phenomenon soon became known as the “ozone hole” (Stolarski et al., 1986; Newman et al., 2022). Paul attended a workshop on the subject in Boulder in early 1986, where he learned about the theories put forward to explain the phenomenon, most notably the idea by Solomon et al. (1986) of chlorine activation on the surface of stratospheric ice particles through the reaction

$$HCl + ClONO_2 \longrightarrow Cl_2 + HNO_3 \tag{14}$$

Already on the flight back to Germany and together with F. Arnold after his return, he developed the idea that in the cold Antarctic stratosphere, heteromolecular condensation of H_2O and HNO_3 occurs (well above the threshold temperature for the formation of pure ice particles), which leads to the formation of HNO_3—H_2O aerosol particles in the stratosphere and, consequently, to a strong depletion of HNO_3 (and thus NO_x) in the gas-phase. (The idea of heteromolecular condensation of H_2O and HNO_3, as well as the removal of NO_x from the gas-phase had also been independently developed by Toon et al. (1986).) The condensation of HNO_3 further leads to an increase in aerosol surface area and thus to an accelerated rate of heterogeneous reactions like reaction (14). The paper on this mechanism was published at the end of 1986 (21).

The ideas of Crutzen and Arnold (21) were an important contribution to the explanation of the ozone hole (albeit not the only one, see reference 30). Today, much more is known, for example that further heterogeneous reactions are important (like $HCl + HOCl \longrightarrow Cl_2 + H_2O$, reference 25). It has further been established that supercooled $H_2SO_4/HNO_3/H_2O$ droplets exist in the stratosphere with important consequences for the microphysics of polar stratospheric clouds and heterogeneous reactions (see e.g. references 28, 33 and Hanson et al., 1994; Solomon, 1999; Shi et al., 2001).

Even in the absence of low temperatures, H_2SO_4/H_2O particles always exist in the lower stratosphere (Junge et al., 1961); these particles are formed from stratospheric SO_2. Direct injections by large volcanic eruptions are one (intermittent) source of stratospheric SO_2. The other source is oxidation of OCS, which is produced at the earth's surface but reaches the stratosphere; this mechanism was also proposed by Paul (10) and soon confirmed by measurements that showed atmospheric concentrations of OCS of approximately 500 ppt[3] (Sandalls and Penkett, 1977).

6.3 The Montreal Protocol

A large number of scientists worked on the impact of chlorine released to the atmosphere as a result of human activities on stratospheric ozone and in particular on the Antarctic ozone hole. This research led eventually to the Montreal Protocol in 1987 and its adjustments and amendments in the following decades (WMO, 2018). Paul made substantial contributions to this research (e.g., references 7, 21, 25, 28, 33). The Montreal protocol is one of the most important environmental treaties agreed on so far and is today ratified by all member states of the United Nations; it is expected to lead to the eventual phase-out of all ozone depleting substances worldwide and it led to the slow decline of the atmospheric halogen loading that is observed today (WMO, 2018).

In a model study Newman et al. (2009) investigated what might have happened had the Montreal Protocol never been signed. In this case, a continued increase in ozone depleting substances in the atmosphere would have occurred at a rate of 3% per year. In such a "world avoided" scenario, large ozone depletions in the polar region become year-round phenomena (rather than just seasonal phenomena as is currently observed) and tropical lower stratospheric ozone collapses to near zero by about 2060. In response to such ozone changes, ultraviolet radiation at the ground also increases substantially (Newman et al., 2009). Paul's research forms part of the backbone of the science by an active group of researchers that fostered policy action such as the Montreal protocol (Solomon, 2021b); it therefore helped avoiding the massive ozone losses described by Newman et al. (2009) in their "world avoided" scenario. Instead today the emergence of a healing of the Antarctic ozone layer is observed (e.g., Solomon et al., 2016; WMO, 2018).

And Paul emphasised that things could have been much worse; he wrote in 1996: "Noting that nobody had worried about the atmospheric consequences of the release of Cl or Br before 1974, I can only conclude that we have been extremely lucky, which shows that we should always be on our guard for the potential consequences of the release of new products into the environment" (from reference 30).

7 "Nuclear winter"

The involvement of Paul Crutzen in "nuclear winter" studies may be explained (30) by his research interests both into the effects of NO_x on stratospheric ozone (section 3) and in biomass-burning (section 5.3). His work on "nuclear winter" started in 1981, when he was asked by the editor of Ambio to contribute to a special issue on the environmental consequences of a major nuclear war. His initial thought was that he would make an update on model predictions of the destruction of ozone by the NO_x that would be produced and carried up into the stratosphere by large nuclear explosions (an issue already discussed

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[3] Parts per trillion, a measure of the molar mixing ratio.

much earlier, 8). In the late seventies and early eighties, atmospheric models had evolved into two-dimensional zonally averaged models, which covered the atmosphere from pole to pole and where transport was represented by a combination of advection terms and a two-dimensional matrix of eddy diffusion coefficients – the zonally averaged transport of such models approximated a diabatic circulation (e.g., Brasseur, 2019).

John Birks (who spent a sabbatical at MPI Mainz at that time) joined him in this study (see also Fishman et al., 2022). In their first set of calculations, to their surprise, no stratospheric ozone depletion resulted from the nuclear explosions. The reason was that the first scenario they used implied that all explosions were less than one megaton (in equivalent weight of conventional explosive), which, as they found, do not reach the stratosphere. When the scenario was widened, to also include explosions of a few megatons each, substantial stratospheric ozone depletion was indeed predicted.

However, Paul and John Birks soon realised that the direct impacts of possible nuclear explosions were much stronger (19). Such possible nuclear explosions should cause large amounts of smoke emissions (as well as NO_x and hydrocarbon emissions) from fires, which would reach the middle and higher troposphere. The resulting absorption of sunlight by the black smoke could lead to darkness and strong cooling at the earth's surface. Further, the black smoke leads to a heating of the atmosphere at higher elevations, thus creating rather atypical meteorological and climatic conditions that have substantial environmental consequences (references 19, 20).

The term "nuclear winter" to describe the phenomenon was coined by the TTAPS (Turco, Toon, Ackerman, Pollack and Sagan) group in their subsequent paper that expanded on the climate effects of the injections of large amounts of soot and dust into the atmosphere as the result of a nuclear war (Turco et al., 1983). Numerous subsequent studies have confirmed that a large-scale nuclear war would cause severe climate effects, including a study by the U.S. National Academy of Sciences (1985); severe climate effects would also be caused by regional nuclear conflicts (Mills et al., 2008).

Although Paul did not count the nuclear winter idea among his greatest scientific achievements, he was convinced "that, from a political point of view, it is by far the most important" (30). He stated that this idea "highlights the dangers of a nuclear war and convinces me that in the long run mankind can only escape such horrific consequences if nuclear weapons are totally abolished by international agreement" (from reference 30).

8 Atmospheric chemistry and climate change

Atmospheric chemistry and climate change are linked; CH_4 has an important chemical sink in the troposphere (reaction with OH, section 5.1) and the main atmospheric sink of N_2O is photolysis in the stratosphere (section 4.3). Likewise, ozone concentrations throughout the atmosphere are determined by atmospheric chemistry and are influenced by human activities. Finally, atmospheric chemistry affects the nature and the concentrations of cloud-condensation nuclei and thus their impact on cloud properties.

These facts led Paul Crutzen to conduct studies on reactions taking place in and on cloud droplets and tropospheric aerosol particles, which result in the removal of NO_x and reduce OH and O_3 in air masses influenced by clouds (23, 26). Another topic of the work of Paul was the impact of (natural) halogens on the formation of cloud-condensation nuclei and on ozone in the marine boundary layer (31).

The most important atmospheric greenhouse gases are CO_2, CH_4, and N_2O, the effective radiative forcing in 2019 (relative to the year 1750) for CO_2 is 2.16 $W\,m^{-2}$, for CH_4 is 0.54 $W\,m^{-2}$ and for N_2O is 0.21 $W\,m^{-2}$ (Arias et al., 2021). Further, tropospheric ozone is a significant greenhouse gas (16), which interacts with terrestrial infrared radiation in the 10 μm region (an atmospheric window region); the effective radiative forcing in 2019 (relative to the year 1750) of ozone (where the forcing is dominated by tropospheric ozone) is 0.47 [0.24 to 0.71] $W\,m^{-2}$ (Arias et al., 2021).

Of greatest importance to radiative forcing are any changes in ozone in the tropopause region and in the tropics and subtropics (Riese et al., 2012). The latter point underlines the importance of observations of tropospheric ozone in the tropics and subtropics, where the atmospheric oxidation efficiency is highest, where high ozone concentrations are observed over the polluted regions of the continents and where observations are particularly sparse (30).

Moreover, emissions of CO_2 and N_2O caused by human activities are not independent. The conversion of biomass to biofuel will allow some emissions of CO_2 (caused by the use of fossil fuel) to be saved, but this biofuel production will lead to some additional N_2O emissions caused by the use of fertilisers (see also section 4.3). Paul conducted an analysis of these compensating effects (49) finding that the production of commonly used biofuels may contribute as much or more to global warming through these additional N_2O emissions than the cooling by the fossil fuel savings.

A lot of Paul's work on the link between atmospheric chemistry and climate change was done in collaboration with his friend Veerabhadran Ramanathan (e.g., 16, 39, 40), who works at Scripps Institution of Oceanography, University of California San Diego in La Jolla, USA. Paul was a regular visitor at Scripps and between 1992 and 2008 served as distinguished professor (part-time) at Scripps in La Jolla.

9 Geoengineering

In 2006 Paul Crutzen wrote an essay (47) entitled "Albedo enhancements by stratospheric sulfur injections: a contribution to resolve a policy dilemma?" In this essay he stated that "the usefulness of artificially enhancing earth's albedo and thereby cooling climate by adding sunlight reflecting aerosol in the stratosphere [. . .] might again be explored and debated . . . " (47). This topic was not entirely new (e.g., Budyko, 1977; Dickinson, 1996) and even Paul himself had used the term "geoengineering" earlier (in 2002, reference 41). Further, this was not a single essay, but rather a special section in *Climatic Change* consisting of six papers in total (56). The essay by Paul had a profound influence; it initiated a large number of studies on geoengineering with many different facets.

Today it is accepted that there are two fundamentally different kinds of geoengineering: (a) actively removing CO_2 from the atmosphere, and (b) cooling the earth by either reflecting more sunlight into space (the original proposal in reference 47) or by modifying cloud properties. The number of scientific publications on all aspects of climate engineering increased very strongly after the publication of the special section of *Climatic Change* in 2006 (56). In an interview in 2013, Paul supported continuing research on geoengineering, but doubted that sunlight-reflecting aerosol would ever be injected into the stratosphere (Möllers et al., 2015). His position (like on other occasions) was that predictions on the effect of geoengineering and the possible environmental consequences should be reliable, before application can be considered.

Soon after the publication of Paul's essay, there was an assessment by the Royal Society (Shepherd, 2009), and many other international assessment reports and research initiatives followed (reference 56 and references therein); the most recent assessment is by the National Academies of Sciences, Engineering, and Medicine (2021). Geoengineering has also been addressed in the frame of the IPCC, and one should particularly emphasise the geoengineering model intercomparison project (GeoMIP, Kravitz et al., 2011).

Clearly, geoengineering would have a lot of unintended side-effects, one important example is that an intentional injection of sulphur into the stratosphere might enhance stratospheric ozone depletion in general and lead to a slower recovery of the Antarctic ozone hole in particular (e.g. Tilmes et al., 2021). Indeed, the most recent (2022) "WMO Scientific assessment of ozone depletion" contains for the first time a chapter on the question of the impact of stratospheric sulphur injections on the stratospheric ozone layer.

The final two sentences of Paul's essay in 2006 should not be forgotten: "... I repeat: the very best would be if emissions of the greenhouse gases could be reduced so much that the stratospheric sulfur release experiment would not need to take place. Currently, this looks like a pious wish" (47).

10 The Anthropocene

Paul Crutzen coined the term "Anthropocene"; it was a spontaneous thought at a symposium of the International Geosphere-Biosphere Program (IGBP) in February 2000 at Cuernavaca, Mexico. He recalls the event: "the chairman spoke of the holocene period the whole time. Suddenly it occurred to me that we were no longer in the holocene. In the holocene period human influence was limited. So I then said: 'We are no longer in the holocene, we are in the anthropocene'. [...] during the coffee break it was the topic of conversation" (from reference 54).

But he soon discovered that E. F. Stoermer (whom he never met) had come up with the term independently and had already used it informally in the 1980s (Möllers et al., 2015; Trischler, 2016). Paul and E. F. Stoermer introduced the concept of the Anthropocene together in a newsletter of the IGBP in the year 2000 (37). Putting forward the hypothesis of a new geological epoch, the Anthropocene, is based on the fact that humanity has become a powerful geological force and will continue to be such a force for thousands of years (41). Paul also suggested that the "Anthropocene could be said to have started in the late eighteenth century, when analyses of air trapped in polar ice showed the beginning of growing global concentrations of carbon dioxide and methane" (reference 41).

The substantial increase of carbon dioxide in the atmosphere, which is driven by the burning of fossil fuel by humans and which started with the industrial revolution is the most notable example of the impact of humanity on nature. But it is clearly not the only one. During the past three centuries, human population has increased tenfold, which has resulted in a huge growth in cattle population (and in an a concomitant rise in atmospheric methane concentrations – the increase is more than 100% today compared to preindustrial concentrations). Fossil fuel burning also causes a release of SO_2 to the atmosphere that is now exceeding the sum of all natural emissions. More nitrogen is now fixed synthetically and applied as fertilisers in agriculture than fixed naturally in all terrestrial ecosystems (see also section 4.3 and reference 11); this has led to an increase of about 20% in atmospheric

N_2O (Fig. 6). Furthermore, humanity has released a type of gas (chlorofluorocarbons) to the atmosphere that has no natural analogues. The release of chlorofluorocarbons to the atmosphere would have caused the destruction of the ozone layer as we know it had no international regulatory measures been taken (see sections 6.2 and 6.3).

These are important examples, but there are many other major and still growing impacts of humanity on the earth system (37, 41, 48, 50, 57, 58). In Paul's own words: "In the Anthropocene, climate change is tightly linked to many of the other grand challenges that we face – including the improvement of air quality, public health, sustainable development, and human well-being" (Al-Delaimy et al., 2020). This underlines the urgency of achieving an effective planetary stewardship (50) in spite of uncertainties in our understanding of the earth system. "In the Anthropocene some risks may be overestimated, while others are underestimated by the best available science at any given time, and human society is constantly forced to make decisions while recognizing that science is incomplete" (Solomon, 2021b).

11 Leadership

Paul Crutzen was always a scientist and never became purely a manager or an administrator. But he was nonetheless responsible for the setting up of many programmes to study and understand chemical phenomena in the atmosphere. He was also available to serve as an advisor of environmental politics; for example he was an advisor of the Enquete commission of the German parliament "zum Schutz der Erdatmosphäre"[4] from 1987 to 1990. And later, as a member of the Pontifical Academy of Sciences, he contributed substantially to formulating Pope Francis' encyclical on climate change (Fishman et al., 2022).

Of particular significance regarding the programmes initiated by Paul was the International Global Atmospheric Chemistry (IGAC) programme of the IGBP (International Geosphere-Biosphere Program) which was set up in 1988 at Dookie college, a small college in Australia, in northern Victoria (see Fig. 8). Following the Dookie meeting, Paul served as the first Vice Chair of the IGAC steering committee from 1988 to 1995 (Chair, R. Prinn) playing a leading role in the establishment of biospheric processes as an essential component of IGAC (Prinn, 1994).

Paul was also instrumental in raising the profile of atmospheric chemistry within earth system science, showing its relevance to the stability of life on earth. This became particularly important following the discovery of the ozone hole and the realisation that large-scale ozone loss in the stratosphere was a definite possibility (see section 6.2).

Although Paul was a theoretician, he always emphasised the importance of observations (in particular atmospheric observations), he once said "observations always come first". There were major observational platforms set up under the influence of Paul; one is the CARIBIC experiment (which is still in operation) where a commercial airliner is used to transport a container equipped with a lot of sensors for measuring individual molecules (42). A large number of molecules including hydrocarbons, halocarbons, nitrogen oxides, CO, ozone, water vapour, etc. is measured on a global scale in the upper troposphere/lower stratosphere in order to improve the understanding of chemical processes and chemical

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[4] In English: "for the protection of earth's atmosphere".

Figure 8: The founding fathers of the International Global Atmospheric Chemistry (IGAC) programme at Dookie college in 1988. From left to right: E. Sanhueza, D. Albritton, R. Duce, I. Galbally, P. Crutzen, D. Ehhalt, P. Buat-Menard, H. Rodhe. (Picture courtesy of Henning Rodhe.)

composition worldwide. He also initiated trace gas measurements on the Trans-Siberian Railroad covering very remote locations (35).

Paul played a key role in the development of the global observing system from space. In the early 1980s he created a group in his department to exploit optical sensors for the measurement of trace gases. Building on the development of Differential Optical Absorption Spectroscopy (DOAS) this group led initially by the late Dieter Perner and John Burrows began in 1984 to develop concepts for passive remote sensing of the atmosphere using solar radiation. In July 1988 this led to the proposal of the SCIAMACHY (SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY), which was subsequently selected for the ESA (European Space Agency) Envisat mission. SCIAMACHY had a wide set of sensors and revealed sources of many important molecules, like methane, sulphur dioxide (SO_2), and nitrogen dioxide (NO_2) (e.g., reference 46). It was typical of Paul that he would contribute to and support such an ambitious pioneering earth observation research mission.

A smaller version of SCIAMACHY, called SCIA-mini, of which Paul was co-Principal Investigator with John Burrows, was proposed for a launch opportunity in late 1988. After descoping to being nadir viewing only, the concept was was selected for launch in 1990 and renamed GOME (Global Ozone Measurement Experiment). A continuing series of instruments resulted from this initiative. GOME, which as the name suggests, had a focus on ozone also delivered measurements of key trace gases; the first global observations of nitrogen dioxide, sulphur dioxide and formaldehyde (HCHO) (34 and references therein). The objective of the GOME instruments was and is to observe the global recovery of O_3 as a result of the measures, enacted by the Montreal protocol (see section 6.3). Paul was also an influential member of the science team of the Halogen Occultation Experiment (HALOE) onboard the Upper Atmosphere Research Satellite (UARS), which measured the vertical distributions of ozone and key trace gases that affect the global ozone distribution (references 27, 29, 36, 38).

One area where Paul had a big impact, but perhaps not so well recognised, was being a founding member of the European Research Council (ERC). He was also deeply involved in the setting up of the Academia Europea. Academia Europea had the intention of highlighting scholastic activities, while the ERC had the intention of funding various research programmes. Paul was instrumental in the launch of the ERC and a highly esteemed member from 2005 to 2008. Paul was always determined to support scientific activities in Europe with a particular focus on cooperation of scientists from different European countries.

Paul was also a key figure in the pioneering work of establishing the new concept of interactive open access publishing; together with Uli Pöschl and Arne Richter he helped to create the journal "Atmospheric Chemistry and Physics (ACP)", which features a public peer-review and an interactive public discussion of submitted manuscripts (51).

In an e-mail on 18 September 2000, A. Richter said that a "meeting of the 'younger and wilder' atmospheric scientists under the lead of Ulrich Pöschl and Paul Crutzen regarding the launch of a new EGS [now EGU] journal on atmospheric chemistry took place on 15 September in Mainz"; this meeting was the birthplace of the new journal ACP (51). The journal ACP was founded in 2000 with Paul as a member of the advisory board. Today, ACP is a very well established scientific journal. In Paul's words: "It has been an amazing journey: over a short period of merely a decade, a novel idea originating from Uli Pöschl and developed by an enthusiastic group of hundreds of scientists, created a new way of scientific publishing and communication, initially covering the fields of atmospheric chemistry and physics. The example has since been followed by many successors in other disciplines, with more to come." (51).

Paul did not shy away from controversies in scientific or even public debate. He was involved in a number of issues that were intensively discussed among scientists and in the public, in particular: supersonic transport (section 4.1), "nuclear winter" (section 7), the ozone hole and the Montreal Protocol (sections 6.2 and 6.3), geoengineering (section 9), and the concept of the Anthropocene (section 10). In all these cases, there was public dispute and intellectual fights between many scientists, but Paul was always on the side of those who eventually prevailed.

12 Overall assessment

The person

Paul Crutzen was a warmhearted, friendly and humorous person; he was always open to everyone. At a conference, or at similar occasions one would have very enjoyable and pleasant evenings with him — and if you were lucky, Terttu would be present as well. He was concerned about the welfare of his fellow human beings; he once said "private things are always important". As busy Paul was with his scientific work, he always had time for his family. All of the Crutzen family have wonderful memories of family gatherings, weekends, holidays, and vacations.

Paul was a very cosmopolitan person, he spoke many languages in addition to his native Dutch. His wife Terttu is from Finland and Swedish became the family language which was spoken at their home. On the occasion of a visit to Jülich (on 15 February 1996) Paul had a conversation with Martin Böhmermann from the Netherlands, who worked in Jülich as a glassblower; of course in Dutch. Böhmermann immediately realised that Paul was from Amsterdam – and not from the richest part of town. Of course Paul's command of English was very good. And the same was true for German. The conversations at the MPI in Mainz were in English, Dutch, German and sometimes in Swedish (depending on the person he talked to). He also spoke French and learned a bit of Finnish.

Paul was travelled a lot and his office was often empty. But he was always accessible – there was always a fax number, where he could be reached. And one could count on a rapid reaction, typically within a day or two, with well thought through comments on any scientific request (be it a paper draft or the chapter of a thesis).

When Paul's mind was set on science, he might forget everything else. It would happen that he asks students or postdocs to come to his home on a Saturday afternoon; after finding them at MPI. At his home there were intense discussions, checking of numbers and examining model results. Paul might even forget to offer tea of coffee – of course Terttu would not. He might also call you in the middle of the night (from the US, the fax machine at MPI was out of paper) asking you to collect several fax messages from the fax at the Meteorology department at the University of Mainz and distribute them to colleagues at MPI. First thing in the morning; these messages were important.

In his scientific work, Paul was very careful with a lot of attention to details. But at the same time he could be rather impatient. He always wanted to see the new results, the new plots or the new simulations immediately. He worked very hard and in a dedicated way; he said once (after discussing extensively the reviewer's comments on the paper of reference 29) "... it is hard work to write a paper – really hard work... Some people claim that they can write a paper in a day – I could not do that".

Paul Crutzen was a very unpretentious person. Susan Solomon remembers a story when as a graduate student, she visited the MPI in Mainz around 1980, and she was at a meeting with a dozen of other scientists where she addressed him as Paul and also disagreed slightly with some technical point he had made. Susan could literally hear the sharp intake of breath from others because she had disagreed. He was oblivious to titles, just smiled and said "ja, you're right". The opposite happened as well; lots of discussions among scientists on a particular issue resulting finally in the conclusion "Paul is right here" – where people had not thought of Paul's point earlier in the discussion.

The scientist

Paul Crutzen was a brilliant scientist – what is particularly remarkable is the breadth of his work. Many great scientists have achieved great advances, but what is particularly impressive about Paul is the number of different topics in atmospheric science to which he made fundamental contributions – these contributions are described above and shall not all be repeated here.

His research interests were broad, encompassing topics in the mesosphere, the stratosphere and the troposphere – with a particular emphasis on the issue of climate change and air quality (12, 14, 16, 32, 39, 40). Further, he initiated the discussion on the question of geoengineering (47) and coined the term "Anthropocene", which he suggested as the geological epoch starting with the industrial revolution. Stating that humanity since that time had had a vast impact on the planet (37, 41).

Paul's work not only had a profound influence on the scientific world, but his work also had an influence on many aspects of environmental politics throughout many countries – to name but a few, the issue of supersonic transport in the stratosphere (section 4.1), the Montreal protocol, (section 6.3), the concept of "nuclear winter" (section 7), and many aspects of air pollution caused by human activities.

Paul's scientific works, especially on the impact of human activities on the atmosphere and climate are influential today and they will continue their influence into the future. They will show the way forward for future work on many scientific questions and likewise for the necessary future policy action to halt the warming of climate.

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There is a lot of biographical material on Paul Crutzen available (Möllers et al., 2015; Lelieveld, 2021; Moortgat et al., 2021; Rodhe, 2021; Solomon, 2021a; Zalasiewicz et al., 2021; Zetzsch, 2021; Fishman et al., 2022, see also references 30, 45, 55); these publications were consulted for the memoir and are recommended for further reading[5].

Data shown in Fig. 6 are courtesy of E. Dlugokencky, D. M. Etheridge, R. Prinn, M. Rubino, R. Wang, and R. Weiss; Law Dome Ice Core data for N_2O may be obtained here `https://data.csiro.au/collections/collection/CIcsiro:37077v1` and data on the recent N_2O atmospheric mixing ratios come from station networks (NOAA/GML: `www.esrl.noaa.gov/gmd/ccgg/trends_n2o/` and AGAGE: `https://agage.mit.edu/data/agage-data`).

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[5] See also the information on the web: `https://www.mpic.de/3864489/paul-crutzen`

AWARDS AND RECOGNITION

Paul J. Crutzen, Mario J. Molina, and F. Sherwood Rowland were awarded the Nobel Prize in Chemistry in 1995 for their ground-breaking work "in atmospheric chemistry, particularly concerning the formation and decomposition of ozone"; further major awards and forms of recognition of Paul J. Crutzen are given below.

Rolex-Discover Scientist of the Year (1984)

Leó Szilárd Award for "Physics in the Publics Interest" of the American Physical Society (1985)

Fellow of the American Geophysical Union (1986)

Tyler Prize for Environmental Achievement (1989)

Corresponding Member of the Royal Netherlands Academy of Arts and Sciences (1990)

Volvo Environment Prize (1991)

Member of the Royal Swedish Academy of Sciences (1992)

Member of the Royal Swedish Academy of Engineering Sciences (Kungl. Ingenjörsvetenskapsakademien IVA) (1992)

Member of the Leopoldina (1992)

Group Achievement Award for outstanding contributions to the success of the HALOE/UARS satellite experiment by NASA Langley Research Center (1994)

German prize for the environment (Bundestiftung Umwelt) (1994)

Member of the National Academy of Sciences (United States of America) (1994)

Global Ozone Award for "Outstanding Contribution for the Protection of the Ozone Layer" by the United Nations Environment Programme (1995)

Member of the "Pontificia Academia Scientiarum" (Pontifical Academy of Sciences) (1996)

"Commandeur in de Orde van de Nederlandse Leeuw" (1996)

Recipient of the "Bundesverdienstkreuz mit Stern" (1996)

Member of the "Accademia Nazionale dei Lincei" (Italy) (1997)

Foreign Member of the Russian Academy of Sciences (1999)

Golden Medal by the Academy of Athens (2003)

Recipient of the UNEP/WMO Vienna Convention Award (2005)

Recipient of the Jawaharlal Nehru Birth Centenary Medal 2006, Indian National Science Academy (2006)

Foreign Member of the Royal Society (ForMemRS) (2006)

International Member of the American Philosophical Society (2007)

Recipient of the "Verdienstorden des Landes Rheinland-Pfalz" Mainz, Germany (2013)

Honorary Member of the “Nationale Akademie der Wissenschaften Leopoldina” (2014)

Honorary Member of the Royal Netherlands Chemical Society (2017)

Lomonosov Gold Medal (2019)

Author Profile

Rolf Müller worked with Paul Crutzen at the "Max-Planck-Institut für Chemie" in Mainz from 1988 to 1995. He gained his PhD under the supervision of Karin Labitzke and Paul Crutzen in 1994 from the Free University of Berlin. He works since 1995 at the Institute of Energy and Climate Research of the Forschungszentrum Jülich. He is also a lecturer at the University of Wuppertal.

No colour photographs of Rolf Müller yet – could be added

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