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Paul J. Crutzen – a pioneer in Earth system science and a founding member of the journal "Atmospheric Chemistry and Physics"

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Abstract. Paul J. Crutzen was a pioneer in atmospheric sciences. At the same time, he was a kind-hearted, humorous person with empathy for the private lives of his colleagues and students, but also with the highest scientific standards for himself and for others. He made fundamental scientific contributions to a wide range of scientific topics in all parts of the atmosphere,
5 from the mesosphere to the stratosphere and to the troposphere near the ground. In particular, he was the first to describe the NO_x-driven ozone depletion cycle in the stratosphere, he developed the first mechanisms for the chemical formation of ozone in the troposphere, he provided key ideas to explain the "ozone hole", and he made fundamental discoveries about the effects of biomass burning on the troposphere. Understanding and addressing the causes of man-made air pollution and climate change was the driving motivation for his scientific work. In his work he did not shy away from challenge and provocation.
10 He pioneered the concept now known as "nuclear winter" and initiated the reopening of the debate on geoengineering. He also brought the term "Anthropocene" to the popular debate. In 2000, Paul was among the founders of the journal "Atmospheric Chemistry and Physics", which was unique at the time in providing public discussion of published preprints, and also what we now call "open access" to published articles. Paul's work on human impacts on atmosphere and climate has had a profound impact on the environmental policies of many countries for decades. In the future, his work will continue to be a guide for
15 generations of scientists and environmental policy makers to come.

1 Introduction

Paul Crutzen was always full of scientific ideas that he pursued and also shared generously with colleagues and students. Furthermore, he was a very hard-working individual; when he focused on a particular scientific problem, he could forget the world around him. Despite all his concentration on science, he always had time for his family, and never forgot how important
20 the private lives of his colleagues and students were. You could always discuss with him events of the day, from political issues to the weather and sports.

Particularly impressive about Paul's scientific achievements is the range of different topics in atmospheric science to which he made fundamental contributions (Müller, 2022; Fishman et al., 2023); a short overview is given below in section 2.2. Paul's research interests included topics in the mesosphere, the stratosphere and the troposphere, with a particular emphasis on the
25 issues of climate change and air quality (e.g., Fishman and Crutzen, 1978; Fishman et al., 1979b, a); in this context, the role of aerosol particles – including black carbon – became a focus of his work (e.g., Lelieveld et al., 2001; Ramanathan et al., 2001). Moreover, he was involved in the first "nuclear winter" studies (Crutzen and Birks, 1982), he initiated the discussion on the question of geoengineering (Crutzen, 2006) and he popularised the term "Anthropocene" as the epoch dating from the commencement of geologically significant human impact on Earth's system (against the resistance of geological institutions,
30 Crutzen, 2002; Crutzen and Steffen, 2003; Crutzen and Müller, 2019; Benner et al., 2021, see also section 4).

Paul was a key figure in establishing the journal "Atmospheric Chemistry and Physics" (ACP). ACP has been a pioneer in transparent peer review since it was founded in the year 2000 (Pöschl, 2004, 2012; Ervens et al., 2023). The special issue "20 years of Atmospheric Chemistry and Physics" of which this paper is a part, and which is celebrating more than 20 years of ACP, contains two papers that are directly related to topics that Paul brought up. These two papers are on nuclear winter
35 (Robock et al., 2023) and on geoengineering (Visioni et al., 2023).

Paul Crutzen himself provided a very good description of his life and of his scientific work in his published Nobel lecture (Crutzen, 1996) on the occasion of the 1995 Nobel Prize in chemistry, which he shared with Mario J. Molina, and F. Sherwood Rowland. Shorter biographical texts are also available (Möllers et al., 2015; Lelieveld, 2021; Moortgat et al., 2021; Rodhe, 2021; Solomon, 2021; Zalasiewicz et al., 2021; Zetzsch, 2021). There are also two more recent and detailed memoirs describing
40 Paul's life and his scientific achievements (Müller, 2022; Fishman et al., 2023) [1]. Finally, there is also a book (Lax, 2018) on the recent history (1959-2000) of the Max Planck Institute (MPI) for chemistry ("Otto-Hahn-Institut") in Mainz, where Paul had worked since 1980; this book also contains a wide range of information on Paul's work during this period.

2   Paul Crutzen: the person and the scientist

2.1   The person

45 Paul Jozef Crutzen was born in Amsterdam on 3 December 1933 and passed away in Mainz on 28 January 2021. He was the son of Anna Gurk and Jozef Crutzen. In Amsterdam, on 14 February 1958, he married Terttu Soininen; Paul and Terttu have two daughters, Ilona and Sylvia, and three grandchildren (Müller, 2022) There is no doubt that Paul was a very hardworking man. He once said "see, this is the life of a scientist, always working". He was very dedicated and demanded the same of his collaborators and students. If the only opportunity to talk science with him was on a Saturday afternoon, you had no choice but
50 to accept his invitation and come to the office. But this intensity also meant that he was always very interested in the work of colleagues and students; you could count on a well-elaborated reply from Paul in a very short time frame to any scientific text you sent him, be it a paper draft, parts of a doctoral thesis or any other kind of text.

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[1]See also the information on the web: https://www.mpic.de/3864489/paul-crutzen
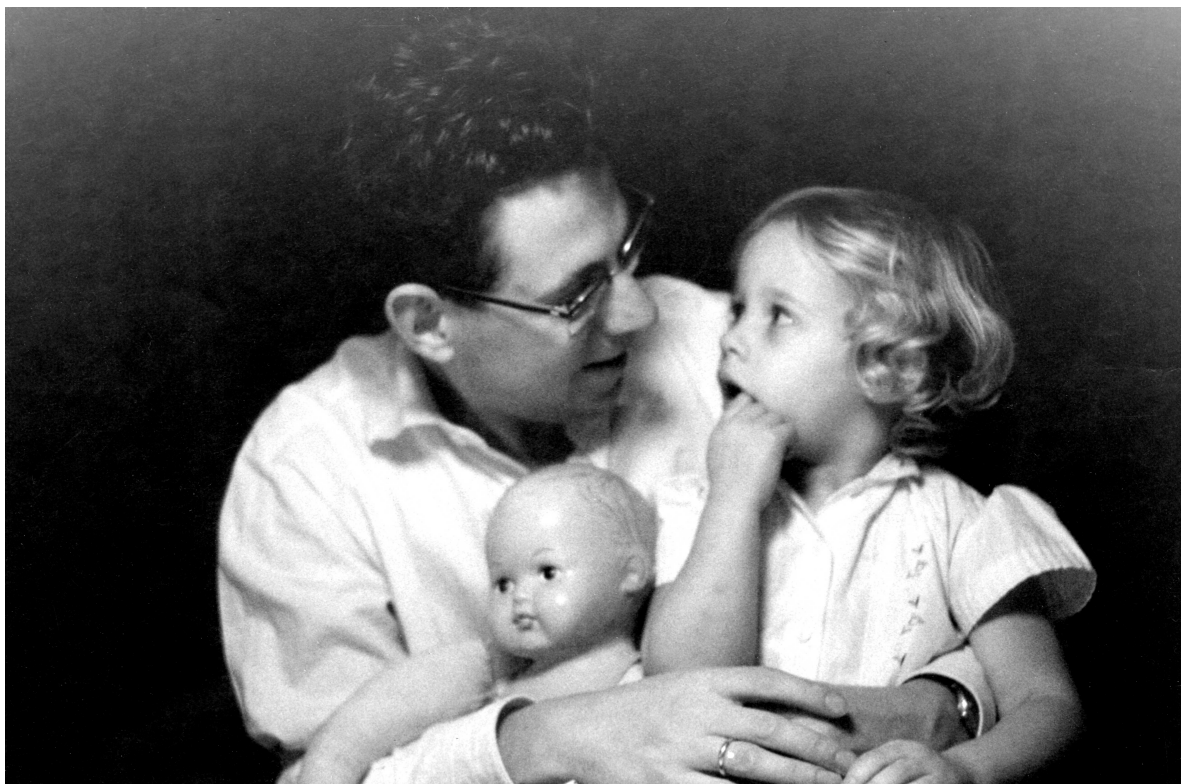
Figure 1. Paul Crutzen with his daughter Ilona in 1961. (Picture courtesy of Ilona Crutzen.)

However, no matter how much Paul concentrated on his scientific work, he always had time for his family (Fig. 1). He himself mentioned that weekends were reserved for family, especially during the time when his daughters were young. The
55 entire Crutzen family has fond memories of family gatherings, weekends, holidays, and vacations with Paul. For his colleagues, Paul was a very pleasant person to be with to whom science was not necessarily the main topic. One could chat with Paul over a coffee or an evening meal on other many topics that he was interested in, such as sports. For example, he was always well-informed about the performance of the Dutch ice skating team, or in football, particularly about the performance of the top team of his hometown (Ajax Amsterdam). Moreover, he followed the football team of the town he lived in (1. FSV Mainz 05)
60 and watched matches live in the Mainz stadium.

2.2 The scientist

Paul's scientific achievements are too numerous and their scope too broad to be covered in detail in this brief note (for more information, see e.g., Crutzen, 1996; Müller, 2022; Fishman et al., 2023). However, stratospheric ozone chemistry in general and NO_x chemistry in particular were what started his scientific work even before his doctoral research (Müller, 2022). He proposed the groundbreaking idea that reactions catalysed by NO and NO_2 control the ozone concentration in the middle
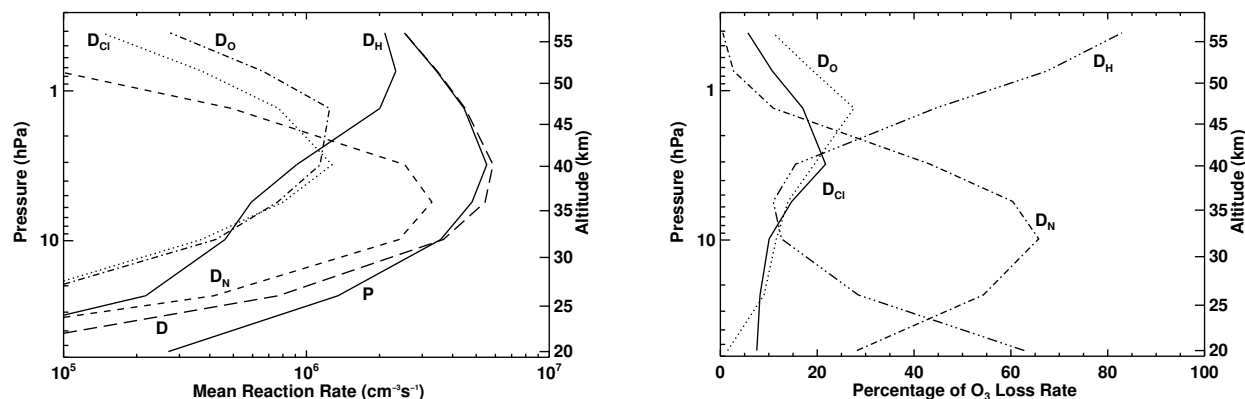
Figure 2. Dependence of various production and destruction reactions on altitude. Left hand panel: the mean reaction rates; right hand panel: the relative importance of the individual contributions to ozone loss in the gas-phase. D_O: Chapman reaction (the reaction $O_3 + O$), D_N: NO_x catalysis (reactions 1 and 2) D_H: HO_x catalysis (by H, OH, and HO_2), and D_{Cl}: ClO_x catalysis (Molina and Rowland, 1974). P: production of odd oxygen (by the reaction $O_2 + h\nu$), D: total ozone destruction. The dominant ozone loss cycle in the stratosphere (D_N) was not known prior to Paul's work (Crutzen, 1970). (Figure adapted from Crutzen et al. (1995) and Grooß et al. (1999); figure courtesy of Jens-Uwe Grooß.)

stratosphere (Crutzen, 1970), according to the following catalytic cycle:

$$NO + O_3 \quad \rightarrow \quad NO_2 + O_2 \tag{1}$$

$$NO_2 + O \quad \rightarrow \quad NO + O_2 \tag{2}$$

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 this mechanism in 1970 was a crucial step towards facilitating a quantitative description of the chemistry of the stratospheric ozone layer; prior to Paul's discovery, the dominant catalytic loss cycle of stratospheric ozone (D_N, through reactions 1 and 2) was not known (Fig. 2). It is now known that D_O: the reaction $O_3 + O \rightarrow 2 O_2$, originally proposed by Chapman (1930), is only a minor sink of stratospheric ozone and that HO_x-induced catalysis only is important close to the tropopause and above about 45 km (e.g., Portmann et al., 2012; Müller, 2021, see also Fig. 2). The recognition that chlorine (D_{Cl}) also catalytically contributes to stratospheric ozone loss came a few years after the discovery of the NO_x-induced cycle (Molina and Rowland, 1974).

Paul was engaged in investigations on the impact of a possible fleet of supersonic planes (and the emissions of NO_x caused by this fleet) on stratospheric ozone (Johnston, 1971; Crutzen, 1972). He also investigated how active nitrogen compounds can enter the stratosphere (Crutzen and Ehhalt, 1977; Müller, 2021). He initiated the first studies on the budget of N_2O in the atmosphere and how it is influenced by human activity (Crutzen and Ehhalt, 1977).

Paul made fundamental discoveries in tropospheric chemistry (e.g., Fishman and Crutzen, 1978; Fishman et al., 1979b; Crutzen and Zimmermann, 1991; Fishman et al., 2023) showing that tropospheric chemical processes provide a much larger source of tropospheric ozone than downward transport of ozone from the stratosphere. This work led him to realise the im-

portance of biomass-burning for the chemistry of the atmosphere (Crutzen et al., 1979; Crutzen and Andreae, 1990). He introduced the notion of "nuclear winter", a concept that the use of nuclear weapons would have global impacts that go much beyond the more obvious direct effects (Crutzen and Birks, 1982). The work on nuclear winter is known today as the hypothesis

80   that widespread firestorms caused by a large-scale nuclear war would inject particles into the stratosphere that block sunlight (Crutzen and Birks, 1982; Robock et al., 2023). While Paul's original intention at the outset of these studies was to explore the impact on stratospheric ozone of nitrogen oxides that might form as a result of a nuclear war (Fishman et al., 2023), this idea has gained considerably broader traction.

After the impact of chlorofluorocarbons (CFCs) on stratospheric ozone was identified (Molina and Rowland, 1974), Paul

85   published a modelling study on this topic in the same year (Crutzen, 1974). In 1985 the ozone hole was discovered by Farman et al. (1985). A year later, Paul – together with Frank Arnold – showed that the formation of stratospheric particles (well above the temperature threshold for ice formation) and the nitric acid uptake into these particles is a crucial aspect of ozone hole chemistry (Crutzen and Arnold, 1986). Heterogeneous chemistry (Solomon et al., 1986) and an ozone loss cycle specific for ozone hole conditions (Molina and Molina, 1987) turned out to be further key processes for explaining the chemical processes

90   responsible for the formation of the ozone hole. Paul suggested that carbonyl sulfide (COS) constitutes the major non-volcanic source for aqueous sulphuric acid aerosol particles in the stratosphere (Crutzen, 1976).

In recognition of the importance of the multi-phase chemistry on atmospheric aerosol particles (Abbatt and Ravishankara, 2023) and the many unknown processes regarding their microphysics, Paul initiated in the early nineties in Mainz a junior research group. The name of the group was: "Heterogene Chemie und Mikrophysik atmosphärischer Aerosolteilchen" [2]. An

95   important starting point of this research was the paper by Luo et al. (1994) reporting on homogeneous and heterogeneous freezing rates of sulphuric acid droplets under stratospheric conditions which has implications for the theory of the formation of nitric acid trihydrate particles in the polar stratosphere. The three authors of that paper are shown in Fig. 3. This photograph was taken by Thomas Koop. At the same time the scene was photographed by A. R. Ravishankara, who also wanted to take a picture of the authors of the paper by Luo et al. (1994).

100   Further work of the junior research group showed the occurrence and importance of a new type of polar stratospheric clouds (type Ib) that consisted of liquid rather than crystalline particles (Carslaw et al., 1994; Koop et al., 1995). It later became clear that these liquid clouds, and not the frozen nitric acid hydrate or ice particles, are the main hosts of heterogeneous chemical reactions of chlorine activation and, thus, polar ozone destruction. In another study (Meilinger et al., 1995), this group showed that the composition and freezing behaviour of the liquid particles depends on small-scale temperature fluctuations in the

105   atmosphere. As it turned out, the smallest droplets reached higher HNO_3 concentrations than larger ones, thus unintuitively increasing the smaller droplets' likelihood to crystallise. When Paul, who was himself rather short in stature, first learned about this result during a discussion, he commented on it with a pinch of self-irony and a big smile: "Never neglect the small ones!".

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[2]in English: "Heterogeneous chemistry and microphysics of atmospheric aerosol particles

Figure 3. Paul Crutzen (right) during a summer school in 1993 together with Thomas Peter (left) and Beiping Luo (middle) at Paestum, Italy. (Picture by Thomas Koop.)

3   Paul Crutzen and the birth of a new journal

For many years (Ehhalt and Ridley, 1996), Paul Crutzen has been an editor of classical journals (*Tellus* and the *Journal of Atmospheric Chemistry*). In 2000, however, Paul was also key in the pioneering work of establishing a new, unique concept of scientific publishing: public discussion of preprints and open access publishing. Together with Uli Pöschl and Arne Richter, Paul helped founding the journal "Atmospheric Chemistry and Physics (ACP)", published by the European Geophysical Society (EGS, which is now the European Geosciences Union (EGU)). ACP was the first journal that featured a public peer-review and an interactive public discussion of submitted manuscripts (Dingwell et al., 2011; Pöschl, 2012). The first paper was submitted to ACP in 2001. The ACP concept is now well established among EGU journals and over the years many newly established journals have followed this example (and future journals will continue to do so, Ervens et al., 2023).

In an e-mail of 18 September 2000, A. Richter wrote 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 journal on atmospheric chemistry took place on 15 September in Mainz"; this meeting was the birthplace of the new journal ACP (Dingwell et al., 2011). The journal

120 ACP was founded in 2000 with Paul as a member of the advisory board. At that time the development and success of ACP
could hardly have been foreseen.

Today, ACP is a very well established and highly ranked scientific journal. Starting with only 7 published papers in 2001
(34 in 2002 and 158 in 2003) the number of papers published by ACP increased steadily until 2010, when more than 800
papers were published. In recent years regularly more than 800 papers per year appear in ACP (Pöschl, 2012; Ervens et al.,

125 2023). 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." (Dingwell et al., 2011).

4 Anthropocene

130 With the Anthropocene concept (Crutzen, 2002; Crutzen and Steffen, 2003; Crutzen and Müller, 2019; Benner et al., 2021),
Paul expressed his insight that humanity is indeed changing the planet as a whole and should take responsibility for its de-
velopment. He actively advocated this concept until recently (Fig. 4). The Anthropocene concept also led to the development
of the iconic "great acceleration" figures (see e.g., Fig. 2 in Fishman et al., 2023), increases in population, greenhouse gases,
fertiliser consumption, and many other signatures of human impact on the Earth system since the industrial revolution (Steffen

135 et al., 2007).

In 2009, the Anthropocene Working Group (AWG, Fig. 4) was established within the Subcommission on Quaternary Stratig-
raphy as an interdisciplinary research group dedicated to formalising the Anthropocene as the current geologic time scale and,
more generally, to studying the Anthropocene as a geological time unit. The term Anthropocene became popular after Paul
Crutzen proposed it spontaneously at a conference in 2000 to assign the current epoch (Benner et al., 2021; Müller, 2022;

140 Fishman et al., 2023). Today, the evaluation of the Anthropocene as a formal unit in the geological timescale continues (Ellis
et al., 2016; Zalasiewicz et al., 2017; Luciano, 2022; Fishman et al., 2023).

5 The impact of Paul Crutzen on atmospheric chemistry and physics

Paul Crutzen had very broad scientific interests and an enormous influence on science; his interests covered practically the entire
atmosphere from the ground to the mesosphere. The body of Paul's scientific work is too extensive and broad to be covered

145 here, but some examples have been given. At the same time, the appreciation of Paul's personality would be incomplete without
acknowledging his interest and care for the private lives of people around him, especially his family.

On top of his contribution to science, he was also a key figure in the development of an entirely new approach to scientific
publishing that began with the journal Atmospheric Chemistry and Physics (ACP). When ACP was founded in 2000, it was
unique in featuring public discussion of published preprints and, furthermore, open access to finally accepted and published

150 papers. The 20st anniversary of this journal is celebrated in this special issue. Paul's legacy is honoured in ACP in form of the

Figure 4. Meeting of the Anthropocene Working Group (AWG) at the Max Planck Institute for Chemistry in March 2017. The people on the picture, clockwise from bottom left to bottom right: Franz Mauelshagen, Institut für transformative Nachhaltigkeitsforschung (IASS) Potsdam; Colin Waters, University of Leicester and AWG; Jürgen Renn, MPI für Wissenschaftsgeschichte, Berlin; Bernd Scherer, Haus der Kulturen der Welt (HKW), Berlin; Jos Lelieveld, MPI für Chemie, Mainz; Reinhold Leinfelder, Freie Universität Berlin and AWG; Davor Vidas, Fridtjof Nansen Institut, Oslo and AWG; Mark Williams, University of Leicester and AWG; Christoph Rosol, HKW und MPI für Wissenschaftsgeschichte, Berlin; Mark Lawrence, IASS, Potsdam; Susanne Benner, MPI für Chemie, Mainz; Jan Zalasiewicz, University of Leicester and AWG; Astrid Kaltenbach, MPI für Chemie, Mainz; Uli Pöschl, MPI für Chemie, Mainz; and Paul J. Crutzen, MPI für Chemie, Mainz and AWG. (Picture by S. Schweller, MPI for Chemistry).

ACP Paul Crutzen publication award, which was created to recognise an outstanding publication in ACP. The first prize was awarded in 2021.

Paul's work not only had a profound impact on the scientific world, but also influenced the environmental politics of many countries. His scientific work will continue to provide guidance for the evolution of science. Likewise, his ideas will continue

155 to have a strong influence on future global policies needed to halt the warming of Earth's climate and the destruction of our planet as we know it.

Data availability. not applicable

Author contributions. U.P., T.K., T.P., K.C., and R.M. all contributed to putting together the material for this paper and to writing the manuscript.

160 *Competing interests.* K.C. and R.M. are editors of ACP; T.K. and U.P. are members of ACP's advisory board.

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