Review of Müller et al. (2023), Paul J. Crutzen – a pioneer in Earth system science and a founding member of the journal "Atmospheric Chemistry and Physics"

This is a great paper and certainly should be published. But Paul's role in nuclear winter is incorrect, and he only contributed to one proposed climate intervention scheme, injecting SO_2 into the stratosphere to create an aerosol cloud, not to geoengineering in general. And we now use climate intervention and not "geoengineering" to refer to this topic, and Paul's paper used "climate intervention" in the title.

Also, all the praise for *ACP* needs to be tempered with a big problem it created, that of Discussions papers that are not peer-reviewed, but have DOIs and end up being referenced. Please see below for details and also respond to the 8 comments in the attached manuscript.

It is not correct that Paul Crutzen "pioneered the concept now known as 'nuclear winter.'" But his work on smoke from fires after nuclear war (Crutzen and Birks, 1982) did lead directly to the theory of nuclear winter calculated by other scientists (Turco et al., 1983; Aleksandrov and Stenchikov, 1983; Robock, 1984; Covey et al., 1984). While Crutzen and Birks (1982) pointed out that there would be fires after nuclear war which would produce so much smoke that it would be dim at Earth's surface, hence the title "Twilight at Noon," that paper never mentioned how surface temperature would change. As John Birks told me last year, they studied tropospheric smoke, not stratospheric, and they knew that the reduction of sunlight would be balanced by heating by absorption of sunlight by the black smoke. It took Turco et al. (1983) and Aleksandrov and Stenchikov (1983), soon followed by Robock (1984) and Covey et al. (1984) to calculate the surface temperature and explain it as nuclear winter. But of course, they were inspired by Crutzen and Birks.

Crutzen and Birks (1982) write about a lifetime for the smoke of weeks and maybe months, and that the smoke would be removed by rain and dry deposition ("Alternatively these reactions begin to occur after an appreciable fraction of the aerosol loading of the atmosphere has diminished because of removal of the particulate matter by rain or dry deposition."). In fact, they say, "Our model does not predict significant stratospheric ozone depletion" for the scenario they studied, but they did write, "Finally, we may point out that there is a possibility that even a nuclear war according to Scenario I, in which most NO_x is deposited in the troposphere, may cause ozone depletions in the stratosphere, if the hot fires in the oil and gas production regions become so powerful that the fire plumes penetrate into the stratosphere. Another means of upward transport may occur when the heavy, dark aerosol layer, initially located in the troposphere, is heated by solar radiation and starts to set up convection and wind systems which will transport an appreciable fraction of the fire effluents into the stratosphere. These speculative thoughts may be pursued further with currently available general circulation models of the atmosphere."

Crutzen and Birks (1982) make no conclusions about the impacts on climate on nuclear war, and did not think it likely. They only speculate about it and say the answers must await further work. They write, "It may be possible to test the impact of nuclear war on climate with this [referring to Jim Hansen's recent work] and similar models when these are supplied with reasonable estimates of the trace gas and aerosol composition of the earth's atmosphere. Whether the induced perturbation in the climate system could lead to longer lasting climatic changes will, however, be difficult to predict. In fact, it may seem unlikely that it will take place. The Krakatoa volcanic eruption of 1883 injected quantities of aerosol into the atmosphere comparable to those which would be caused by a nuclear war, and global mean temperatures were affected for only a few years (1). Still, we must be cautious with a prediction as the physical characteristics of the aerosol produced by volcanos and fires are different, and much is still unknown about the fundamentals of climatic changes. For instance, we may ask questions such as whether the earth's albedo would be substantially altered after a nuclear war and thus affect the radiation balance or whether the deposition of soot aerosol on arctic snow and ice and on the glaciers of the Northern Hemisphere might not lead to such heavy snow and ice melting as to cause an irreversible change in one or more important climatic parameters."

Paul did reopen the debate about geoengineering (now known as "climate intervention"), but only one particular kind. He only discussed solar radiation modification, and not carbon dioxide removal. In fact, he only discussed one proposed scheme, "albedo enhancement by stratospheric sulfur injections."

Lines 149-150: You should also mention the problem that open access to ACP preprints creates. They are assigned a DOI, and I have seen many submitted and even published papers that reference Discussions papers. Sometimes these are rejected papers that should never be referenced, and all the others are not peer-reviewed and have issues that were resolved in the final versions. Just saying how wonderful ACP is while ignoring this problem is disingenuous.

I reproduce below two photos of the instructors at "Governing Climate Engineering – A Transdisciplinary Summer School," Max-Planck-Institute for Comparative Public Law and International Law, Heidelberg, Germany, July 12-16, 2010. Unfortunately, there were no women, and you might want to comment on that. Nowadays many women are working on this topic. I can identify the Americans there, David Keith, Phil Rasch, and Alan Robock, and also Tom Peter and, of course, Paul. I am sure you can identify the others, and you might want to consider using one of these photos in the paper. They illustrate Paul's continuous interest in this important research area that he stimulated. They are my photos and you are free to use them. If you want high-resolution versions, please let me know.

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Review by Alan Robock

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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, 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. 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 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 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.

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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 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, 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 (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 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

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 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.

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

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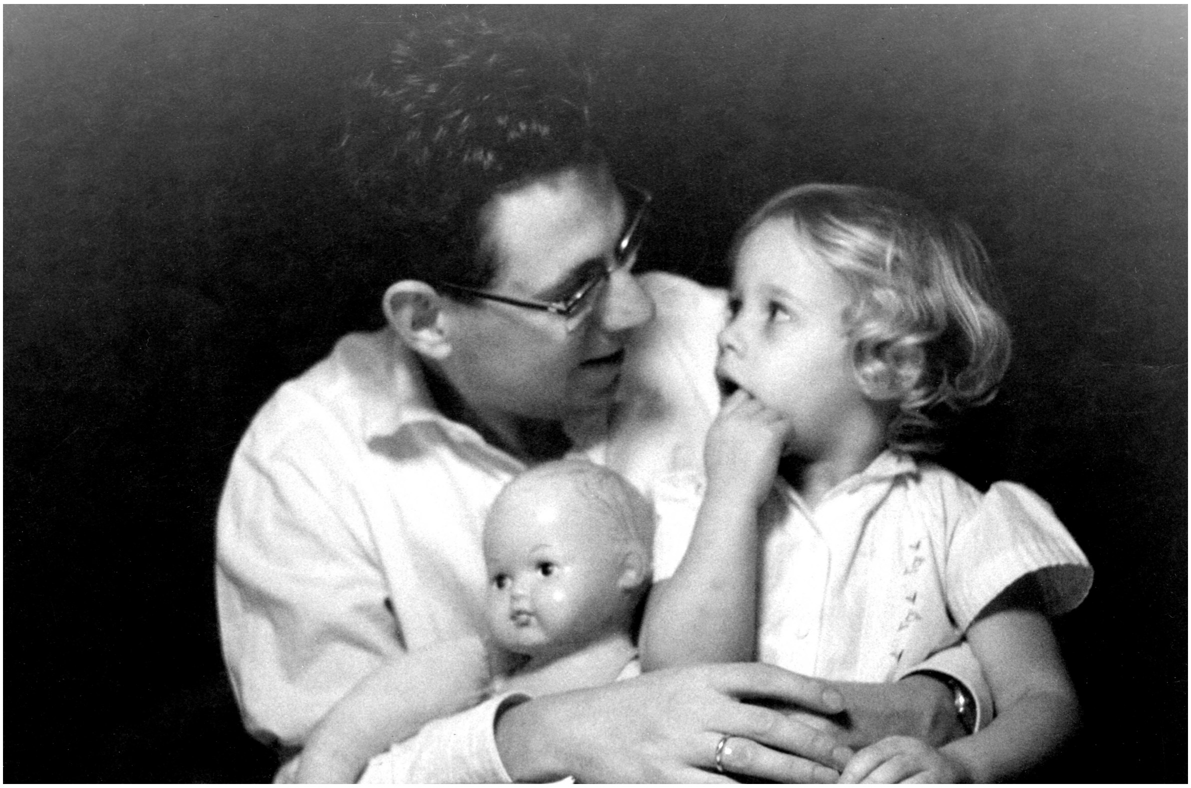

Figure 1. Paul Crutzen with his daughter Ilona in 1961. (Picture courtesy of Ilona Crutzen.)

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 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) 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

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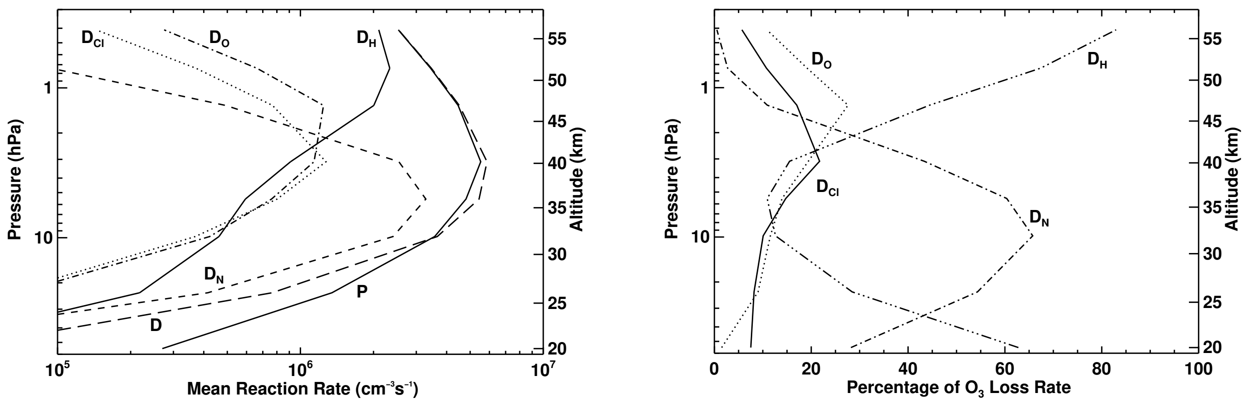

Figure 2. Dependence of various production and destruction reactions on altitude. Left hand panel: the mean reaction rates; right hand panel: the relative importance of the individual contributions to ozone loss in the gas-phase. D_O: Chapman reaction (the reaction $O_3 + O$), D_N: NO_x catalysis (reactions 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:

$$NO + O_3 \rightarrow NO_2 + O_2 \quad (1)$$

$$NO_2 + O \rightarrow NO + O_2 \quad (2)$$

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-

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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 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 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 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 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).

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 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!".

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

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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

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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., 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

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 development. 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 et al., 2007).

In 2009, the Anthropocene Working Group (AWG, Fig. 4) was established within the Subcommission on Quaternary Stratigraphy 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; 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 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 papers. The 20st anniversary of this journal is celebrated in this special issue. Paul's legacy is honoured in ACP in form of the

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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).

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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 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.

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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