

## A Tribute to Paul Crutzen (1933–2021)

### The Pioneering Atmospheric Chemist Who Provided New Insight into the Concept of Climate Change

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**ABSTRACT:** Paul Crutzen received his doctorate in meteorology from the University of Stockholm in 1968 and was awarded the Nobel Prize in Chemistry in 1995. In addition to chemistry and atmospheric science, however, the breadth of his accomplishments has also been recognized by biologists, Earth system scientists, and geologists. This tribute provides some insight into Crutzen's career and how it contributed to so many scientific disciplines. In addition, we offer a road map showing how these diverse contributions were woven together over the course of more than five decades of research. The citation for the 1995 Nobel Prize reads that it was given for "work in atmospheric chemistry, particularly concerning *the formation and decomposition of ozone*." The inclusion of the wording "*formation ... of ozone*" applies only to him among the three laureates (Crutzen, Mario Molina, and F. Sherwood Rowland). His research on tropospheric chemistry led to seminal studies of tropical biomass burning, which eventually evolved into the concept later known as "nuclear winter," a topic in the forefront of far-ranging popular discussions in the 1980s. Last, Crutzen's proposal for the emergence of the "Anthropocene" as a new geological epoch that would terminate the 11,700-yr-old Holocene is considered by the Earth system science community to be the most pronounced trademark of his remarkable career. Crutzen also received American Meteorological Society's Battan Award for his coauthorship of *Atmosphere, Climate, and Change*, recognized by the organization as the best book for general audiences. In the later years of his career, as a member of the Pontifical Academy of Sciences, Crutzen was a key player in the formulation of *Laudato Si'*, Pope Francis's encyclical on climate change, which was released in advance of the Conference of Parties (COP 21) meeting that announced the formulation of the Paris Climate Accords in 2015.

**KEYWORDS:** Anthropogenic effects/forcing; Chemistry, atmospheric; Climate change; Ozone; Biomass burning

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**P**aul Crutzen received both his bachelor's and master's degrees in meteorology from the University of Stockholm while he was employed as a computer programmer at the university's Meteorological Institute (MISU). He started his studies in 1959 at the age of 25, shortly after MISU's founding director, Carl-Gustav Rossby, died and Bert Bolin had taken over its leadership. By 1963, he had completed enough coursework to earn the equivalent of a master's degree (Filosofie Kandidat) and was encouraged to pursue a doctorate degree. Although the University of Stockholm was still a world leader in the field of dynamic meteorology because of Rossby's influence, and was likewise leading the way in the study of acid rain, a field of expertise headed by Bolin, Crutzen's research topic evolved from the work he did as a programmer when an American visitor, James Blankenship, worked with Georg Witt, another one of MISU's scientists. Witt's expertise was the study of noctilucent clouds, which form in the upper stratosphere and mesosphere at altitudes near 85 km.

As part of one of his programming jobs, Blankenship needed to calculate the vertical distribution of the allotropes of oxygen (O, O<sub>2</sub>, and O<sub>3</sub>) through the stratosphere, mesosphere, and lower thermosphere (Blankenship and Crutzen 1966). Crutzen continued and expanded this modeling work as a topic for his Ph.D. dissertation under the direction of Bolin and Witt, two distinguished scientists whose fields of interest focused on diverse regions of the atmosphere: from the very bottom where Bolin's work eventually evolved into understanding the interaction of the biosphere and the atmosphere [Bolin would become the founding director of the International Geosphere-Biosphere Program (IGBP)] whereas Witt would continue to make contributions to studies of the mesosphere and thermosphere.

To help Blankenship on his problem, Crutzen needed to learn stratospheric chemistry so that the distributions of atomic oxygen (O) and ozone (O<sub>3</sub>) could be simulated; this expanded work formed the basis of his initial dissertation for his Filosofie Licentiat (Ph.D.) in 1968. The understanding of stratospheric chemistry at the time included only oxygen and hydrogen photochemistry (also known as "wet photochemistry"), and the calculated O<sub>3</sub> distribution did not agree well with the observations at the time. Thus, he hypothesized that the inclusion of nitrogen chemistry would likely lead to a calculated ozone distribution that agreed better with the observations. This expanded chemistry became the focus of his advanced doctorate, Filosofie Doctor (D.Sc.) under the direction of Richard Wayne and Sir John Houghton at Oxford University in 1973, after working with Wayne as a postdoctoral fellow from 1969 to 1971 (see "An unusual educational journey from Amsterdam to Stockholm to Oxford" sidebar).

## An unusual educational journey from Amsterdam to Stockholm to Oxford\*

Paul Crutzen was born in Amsterdam ("only 100 meters from the Heineken brewery," he used to brag) in 1933. His father was a waiter (often unemployed) and his mother worked in a hospital kitchen. In 1940, he entered elementary school and the Netherlands were overrun by the German army. The last months of the war, between the fall of 1944 and Liberation Day on 5 May 1945, were particularly horrible. During the cold "hongerwinter" (winter of famine) of 1944/45, there was a severe lack of food and heating fuels. Crutzen would say he would have been several inches taller had he not lived under such harsh conditions during these growth years. Many died of hunger and disease, including several of his schoolmates.

In 1946, he entered middle school, which, in the Dutch educational system, prepared better students for university entrance. However, because of a high fever, his grades in the final exam for entrance into the academic track were not good enough to qualify for a university study stipend. Because he did not want to be a further financial burden on his parents, Crutzen chose to attend a technical school, where he studied civil engineering and construction. This educational track also provided a practicum, which he could live on for a couple of years. From 1954 to February 1958, with a 21-month interruption for compulsory military service in the Netherlands, he worked at the Bridge Construction Bureau of the city of Amsterdam. After marrying Terttu, a Finnish student he had met on a ski vacation in February 1958, they settled in Gävle, a little town about 200 km north of Stockholm, where he had found a government construction job.

Later that year, he replied to a newspaper advertisement from MISU announcing an opening for a computer programmer. Although he had no experience in this subject, Crutzen applied for the job and started his new career in July 1959; he and Terttu moved to Stockholm and he started taking college-level courses for the first time at the age of 25. While working at MISU, he was able to take courses and by 1963, he had obtained enough credits to earn a master's degree. MISU, under the direction of Bert Bolin, had always had a strong interest in environmental problems and had been recognized as a world leader in the ongoing acid rain problem at that time. During this period, he also worked on research headed by Georg Witt, whose interest was in noctilucent clouds and the chemistry and physics of the ionosphere. Working with Witt afforded Crutzen an opportunity to work with a visiting American scientist who was interested in understanding the formation and the distribution of the allotropes of oxygen in the stratosphere and mesosphere (Blankenship and Crutzen 1966). To understand this problem, Crutzen had to solve the equations that controlled stratospheric chemistry that determined the amount of ozone there. Crutzen had always wanted to be an academician so when he decided to pursue his dream and get a Ph.D., he chose to study stratospheric chemistry. Both Witt and Bolin supported this work for his Ph.D., which he received in 1968 where his dissertation was entitled, "Determination of parameters appearing in the 'dry' and the 'wet' photochemical theories for ozone in the stratosphere" (Crutzen 1969) (Fig. SB1).

Throughout his studies, Crutzen never took a chemistry course because he was also employed full time, and the



**Fig. SB1.** Paul Crutzen (center) obtained his Filosofie degree (D.Sc.), in 1973, which is awarded in recognition of a substantial and sustained contribution to scientific knowledge beyond that required for a Ph.D. He is flanked by his examiners, Prof. Richard P. Wayne (left) and Sir John Theodore Houghton (right). (Photo courtesy of Richard P. Wayne.)

laboratory portion of these courses was too time consuming. In 1969, he received a European Space Research Organization postdoctoral fellowship at Oxford University and versed himself in the details of chemical kinetics while cultivating a close relationship with chemist Richard Wayne, focusing on learning the details of additional chemical reactions involving the oxides of nitrogen that were included in his next generation of photochemical models. During his initial Ph.D. research, Crutzen realized that the photochemistry in his model, which included only reactions with oxygen and methane and water vapor (Norrish and Wayne 1965), did not provide the mechanisms needed to simulate the observed ozone distribution in the stratosphere (Crutzen 1970). With the inclusion of nitrogen chemistry, Crutzen also realized that these additional reactions could impact tropospheric processes (Crutzen 1974). The pursuit of understanding the chemistry of the troposphere served as the springboard for hypothesizing the presence of previously undefined phenomena that eventually led to a new vernacular that included the previously undefined terms “nuclear winter” and the Anthropocene.

\*Some of this content is paraphrased from Crutzen’s Nobel lecture (Crutzen 1996).

Crutzen realized that expanding the chemistry to include nitrogen compounds not only impacted stratospheric chemistry, but he also insightfully recognized that a similar set of reactions (e.g., as described in Haagen-Smit 1952) could catalytically produce ozone in the troposphere as well as destroying it catalytically in the stratosphere (Crutzen 1973, 1974). Whether or not ozone was made or destroyed in the atmosphere was critically dependent on the rates of several key reactions that involved reactive hydrogen radicals (H, OH, and HO<sub>2</sub>) and that there was enough uncertainty in the published rate constants at the time that photochemical models could produce a wide range of results.

**Coming down to the troposphere.** The 1995 Nobel Prize in Chemistry was awarded to Paul Crutzen, Mario Molina, and F. Sherwood Rowland (see “Meteorologists as Nobel Laureates” sidebar). Both Molina and Rowland were chemists, where, as mentioned earlier, Crutzen’s doctorate degrees were in meteorology. Even though Crutzen had no formal education in chemistry while at the University of Stockholm, he gained much insight into the field of atmospheric chemical kinetics during the time he spent as a postdoctoral fellow in Richard Wayne’s laboratory at Physical Chemistry Laboratory at the University of Oxford from 1969 to 1971.

A prevailing misconception of the 1995 Nobel Prize for Chemistry is that it was primarily centered on the discovery of the ozone hole in the early 1980s (Farman et al. 1985), several years after the prediction of the effect that chlorofluoromethanes had on ozone depletion described in the seminal paper by Molina and Rowland (1974). However, the citation that accompanies the 1995 Nobel Prize reads, “for their work in atmospheric chemistry, particularly concerning the *formation and decomposition of ozone.*” There is no mention of stratospheric chemistry or the depletion of the ozone layer. A key phrase in this citation, and the one that separates Crutzen from the other two recipients, is the inclusion of the words “formation of ozone.”

As mentioned earlier, Crutzen’s doctoral research focused on the inclusion of reactive nitrogen chemistry on stratospheric ozone. Coming to Boulder, Colorado, in 1975 with a joint appointment at the National Center for Atmospheric Research (NCAR) and at NOAA’s Aeronomy Laboratory, he had recently developed a two-dimensional (latitude–altitude) model of atmospheric chemistry that included oxygen–hydrogen–nitrogen chemistry and parameterized two-dimensional transport (Louis 1974). Several years earlier, his model was a key tool that provided insight into the discussion of the potential impact of supersonic transport airplanes flying in the lower stratosphere (Crutzen 1972; Hildalgo and Crutzen 1977). The model’s vertical domain was from 10 to 55 km and Crutzen realized that a better

## Meteorologists as Nobel Laureates

In 2021, Syukuro Manabe shared the Nobel Prize in Physics for “for the physical modeling of Earth’s climate, quantifying variability and reliably predicting global warming.” Although several scientists had been strongly considered for the Nobel Prize in previous years, no meteorologist or atmospheric scientist had previously received this honor. Perhaps most notable is Vilhelm Bjerknes for his work regarding polar-front theory stemming from his classic paper (Bjerknes 1904); he was nominated 26 times between 1923 and 1945 (a scientist can have more than one nomination each year) for the prize in physics. Sydney Chapman, who is best known for his research that described why the ozone layer existed in the stratosphere was nominated for the physics prize four different times between 1956 and 1963 (nominees are no longer listed after 1966). Although the science of meteorology clearly falls under the purview of physics rather than chemistry, Crutzen’s 1995 co-Laureates (Fig. SB2) were both established chemists in their own right. Rowland’s early research had an environmental focus studying how radioactive material worked its way through the food chain and Molina had established himself in the laboratory as a kineticist measuring reaction rate constants of atmospheric trace gases. He came to the University of California, Irvine, in 1973 as a postdoc for Rowland when they wrote their classic 1974 paper on the possibility of chlorofluorocarbons reaching the stratosphere where they would photolyze to release reactive chlorine and destroy the ozone layer.

Somewhat ironically, both Molina and Rowland concentrated on tropospheric chemistry later in their careers after receiving the Nobel Prize. Rowland’s primary interest was studying the composition of Earth’s atmosphere in remote locations throughout the world, focusing on areas with special conditions, such as burning forests and/or agricultural wastes, or the marine boundary layer in oceanic locations with high biological emissions. His research group collected whole air samples collected on land, ships, and aircraft and were returned to his laboratory at UCI for analysis ([www.faculty.uci.edu/profile.cfm?faculty\\_id=2923](http://www.faculty.uci.edu/profile.cfm?faculty_id=2923)).

Molina and his former wife and long-time collaborator, Luisa Tan Molina, began work on air quality in megacities and eventually formed the Molina Center for Strategic Studies in Energy and the Environment. To steer policy, the Mexico City Project combined unprecedented large-scale field studies of atmospheric chemistry in urban neighborhoods, involving hundreds of international scientists, with in-depth analysis and stakeholder engagement. This work improved the air quality in his beloved home city (McNeill 2020).



**Fig. SB2.** Photo taken at a press conference at the University of Stockholm following the 1995 Nobel Prize ceremony. From left to right: Nobel Laureates Mario Molina, F. Sherwood Rowland, Paul Crutzen with Henning Rodhe, Crutzen’s classmate at the University of Stockholm (courtesy of Henning Rodhe).

lower boundary was needed and that the chemistry below the tropopause required a set of chemical reactions that differed from those that had been incorporated into his stratospheric model (Crutzen 1973, 1974). This task was accomplished by his first American student, Jack Fishman, shortly after Crutzen arrived in the United States (Fishman and Crutzen 1977). Using calculations from this new tropospheric model, Crutzen and Fishman entered the ongoing debate about the origin and budget of tropospheric ozone (Chameides and Walker 1973, 1976; Fabian 1973, 1974), where the atmospheric photochemical modelers' calculations claimed that the process of methane ( $\text{CH}_4$ ) oxidation was a much larger source of ozone than what was being transported downward from the stratosphere through mechanisms such as troposphere–stratosphere exchange (Fabian and Junge 1970). Such transport occurred during the evolution of synoptic-scale events that generated troposphere folds (e.g., Danielsen 1968; Danielsen and Mohnen 1977).

Crutzen realized that the credibility of the atmospheric chemistry models used in the tropospheric ozone controversy was limited by the accuracy of the available measurements (both observational and laboratory derived) used to generate the calculations within the model. Furthermore, he was keenly aware that the results being published were critically dependent on the rates of several key reactions used in these calculations. The models' chemistry is driven by reactions of radicals (i.e., fragments of molecules, such as H, OH, and  $\text{HO}_2$ ), which are generally present in very low concentrations, due to their high reactivities. At the time, none of these reactive species had ever been directly observed in the atmosphere, but the theoretical calculations produced in these models showed that these highly reactive species must be present for the calculations to have any validity.

Being at the Aeronomy Laboratory, Crutzen had access to ongoing chemical kinetic research and was able to use the latest state-of-the-art laboratory measurements to refine his model. Of significant importance was the ability to measure hydrogen radicals directly so that their reactions with longer-lived trace gases could be measured quantitatively rather than being inferred from measuring the decay rate of the longer-lived reactant that was being removed from the system. Specifically, the direct measurement of the hydroperoxyl radical ( $\text{HO}_2$ ) with nitric oxide (NO) was now found to be more than an order of magnitude faster (Howard and Evenson 1977) than had previously been measured. The relevance of this new reaction rate was described in a subsequent paper (Crutzen and Howard 1978). As a result, the relative concentrations of many reactive species significantly altered their roles in atmospheric chemistry, sometimes with surprising results. For example, under certain conditions, the nitric oxide emissions from SST exhaust would not deplete ozone but the revised calculations showed that ozone could be generated in the lower stratosphere.

Using improved reaction rate data also provided an efficient mechanism for generating ozone in the unpolluted (i.e., nonurban) troposphere that had not been considered previously. As a result, these new calculations indicated that carbon monoxide (CO), as well as other products of fossil fuel combustion, were likely the primary sources of photochemically generated ozone (Fishman and Crutzen 1978; Fishman et al. 1979b), and that such sources were several times larger than what was generated from methane oxidation. This connection of understanding the relationship between ozone and CO (which was dominated by anthropogenic fossil fuel use) increased the necessity to better define the global CO budget. The search for a new source of CO, in turn, resulted in the premise that significant sources of carbon monoxide needed to be identified to put the CO budget in balance (Crutzen and Fishman 1977).

***The relationship between CO and tropospheric  $\text{O}_3$ : Predicting the Anthropocene.*** Incorporating the photochemistry initially described in Crutzen (1974) and modified in Fishman and Crutzen (1977), Crutzen oversaw the development of what he called a diagnostic

two-dimensional model of the troposphere to calculate the global distribution of OH. In addition to the meteorological and photochemical parameters, he needed to know the monthly altitude–latitude distributions of temperature, water vapor and ozone, as well as a few other trace gas distributions (e.g., CO and CH<sub>4</sub>) that had been derived from the best available, but very limited, field measurements at the time. The most critical trace gas distribution that needed to be prescribed was the one for ozone. Although the two-dimensional distribution in the Northern Hemisphere was available from a preprint of Chatfield and Harrison (1977), the corresponding distribution in the Southern Hemisphere (SH) had never been published. A novice graduate student, who was staying in Boulder during the summer of 1977 on her way to begin graduate school in California, Susan Solomon, pored through all the available ozonesonde measurements from the World Ozone Data Center (WODC) to develop the ozone distribution in the SH that would be used in these calculations. The resultant OH distribution for the NH was published in Crutzen and Fishman (1977) and then described for both hemispheres in Fishman et al. (1979b).

At the time of these studies in the late 1970s, the understanding of the origin of tropospheric ozone was very much in debate where it had been hypothesized for the first time that photochemistry could play a significant role in the chemistry of the unpolluted troposphere. Earlier studies [prior to the publication of Levy (1971)] had intrinsically assumed that any ozone found in the lower atmosphere outside of polluted urban environs must have originated in the stratosphere (Junge 1962; Fabian and Junge 1970; Fabian 1973; Fabian and Pruchniewicz 1977). When Chameides and Walker (1973, 1976) expanded Levy's (1971) tropospheric OH chemistry to examine tropospheric ozone formation, they concluded that methane oxidation was the dominant source of tropospheric ozone. This premise was refuted in several papers (e.g., Fabian 1974; Danielsen and Mohnen 1977), and the primary argument put forward was that if photochemistry did play the dominant role in the formation of tropospheric ozone, then the highest concentrations should be found at low latitudes, where photochemical activity was most intense. Available observations generally showed just the opposite: concentrations in the tropics were consistently lower than what was found at northern middle latitudes (e.g., Fabian and Pruchniewicz 1977).

An alternative theory highlighted in Fishman and Crutzen (1978) and then expanded upon in Fishman et al. (1979b), showed that the origin of tropospheric ozone, for the first time, was dependent on the presence of a predominantly anthropogenic source, CO, leading to the statement “If significant tropospheric ozone production takes place, it follows that the concentrations of ozone in the lower troposphere in the NH have increased substantially since the inception of the industrial era.” If proven true, then this statement portended the hypothesis put forth in Crutzen (2002) that humankind had greatly altered the composition of the (lower) atmosphere, a primary tenet behind his hypothesis that humankind had entered a new geological epoch, which he referred to as the Anthropocene. The amount of tropospheric ozone data available at the time of the 1978 and 1979 studies, however, was not able to confirm or refute that the onset of the Anthropocene could be identified through the analysis of the existing tropospheric ozone measurements.

Several comprehensive studies describing the tropospheric ozone distribution and its trend were eventually published in the subsequent decades (e.g., Logan 1994, 1999; Volz and Kley 1988) and a synthesis of some of these datasets was published by Marenco et al. (1994), producing the trend shown in Fig. 1, dating back to the late nineteenth century. It should be noted that the tropospheric ozone data from the late nineteenth century and early twentieth century in the Marenco et al. (1994) study had initially been published by Volz and Kley (1988) by recalibrating measurements from the Pic du Midi Observatory using the Schönbein paper measurement technique. These century-old measurements have subsequently been reexamined and the current estimates of these concentrations is believed to be in the

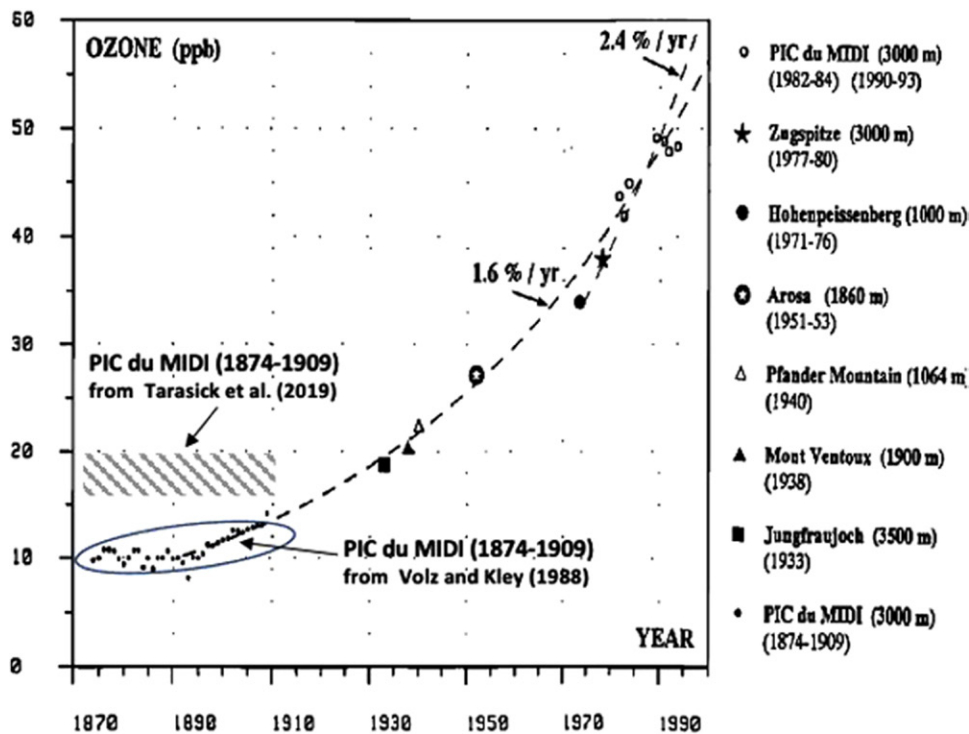


Fig. 1. Tropospheric ozone trend from Marenco et al. (1994) modified with the reanalysis of the Pic du Midi data (1874–1909) described by Tarasick et al. (2019).

15–20 ppb range (Tarasick et al. 2019) rather than ~10 ppb, as reported by Volz and Kley (1988). Even with this adjustment for the  $O_3$  measurements in the late nineteenth and early twentieth centuries, the trend depicted by Marenco et al. (1994) remains valid and resembles what is seen for other longer-lived trace gases such as methane, carbon dioxide, and nitrous oxide, all of which are known to trap outgoing infrared radiation, thus contributing to global warming. Tropospheric ozone is likewise a greenhouse gas and also contributes to global warming, possibly as much as methane (Fishman et al. 1979a; IPCC 2013).

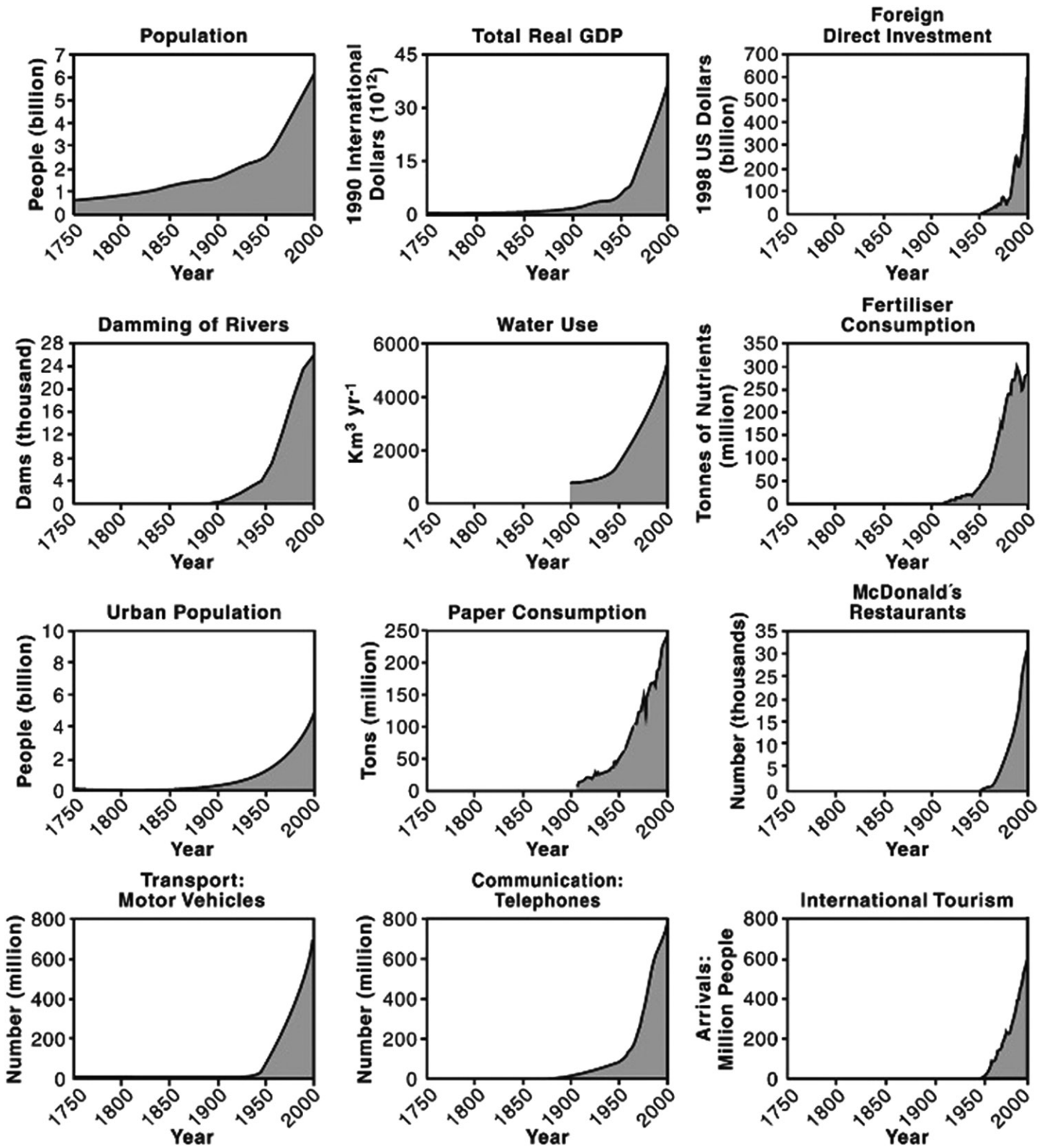
**The “eureka moment” more than two decades in the making.** Tracking back to the 1978 study, Crutzen was likely aware of the general shape of the tropospheric ozone curve shown in Fig. 1, a trend that showed a sharp increase in ozone concentration through the twentieth century, with the rate increasing with time. But Crutzen’s knowledge of how the Earth system was changing spanned many other areas of research in addition to atmospheric chemistry. As a vice chair of IGBP, he was exposed to the most recent research in a wide array of fields, including marine biology, coastal zone dynamics, terrestrial ecology, biodiversity conservation, land-use change, and more. In all these fields, the accelerating, human-driven changes of the twentieth century were evident.

The crucial moment that redefined Earth’s geological history occurred at a meeting in February 2000 of the Scientific Committee of the IGBP in Cuernavaca, Mexico (Zalasiewicz et al. 2021). Crutzen was listening, with increasing exasperation, to the presentation of IGBP’s paleoscience project, a presentation that repeatedly referred to the Holocene epoch (the last 11,700 years of relatively stable, warm conditions that had followed the last ice age), despite the vast array of evidence of the dramatic changes in global environmental parameters in recent decades. His exasperation spilled over into an interjection that we “were no longer in the Holocene but in ... (pausing to try to think of the appropriate word) ... the Anthropocene.” Following up on his Cuernavaca intervention, Crutzen formalized his proposal in his paper, “Geology of Mankind,” which introduced the scientific community to the concept that is now referred to as the Anthropocene (Crutzen 2002). In that paper, he specifically proposed that



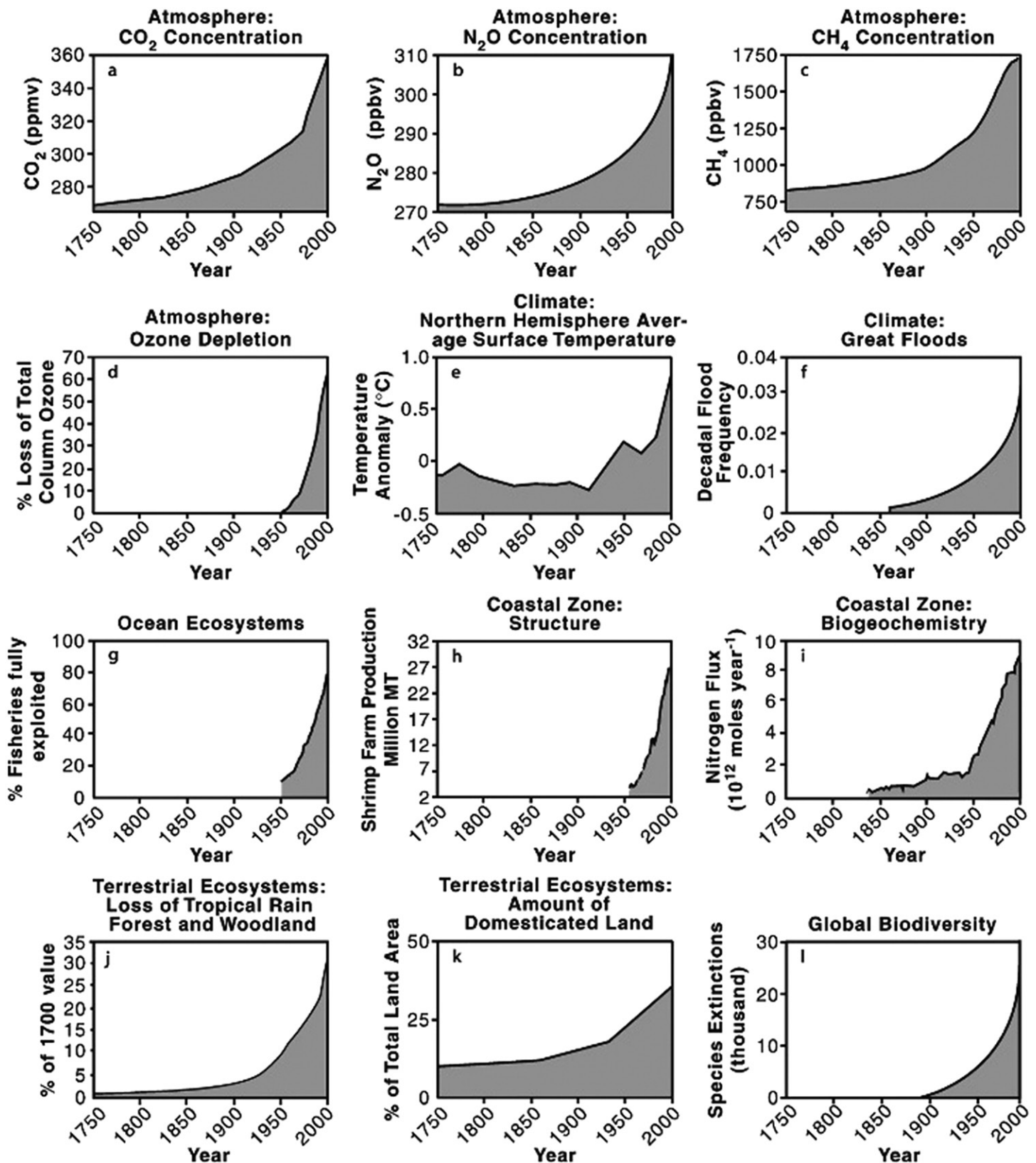
the Anthropocene be formalized at the level of an epoch in the geological time scale, thus terminating the Holocene (the current 11,700-yr epoch).

Crutzen's Cuernavaca interjection also contributed to the development of the iconic "Great Acceleration" figures (Fig. 2), which showed a similar shape to the tropospheric



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Fig. 2. The original Great Acceleration figures showing the (a) increasing rates of change in human activities since the beginning of the industrial revolution and (b) global-scale changes in the Earth system as a result of human activity (Steffen et al. 2004).



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Fig. 2. (Continued).

O<sub>3</sub> measurements in Fig. 1. Earlier versions of the Great Acceleration figures were being developed as part of a major IGBP synthesis project, but these earlier figures spanned only the 1900–2000 time period. Following Crutzen’s (2002) *Nature* paper, in which he suggested that the Anthropocene began with the advent of the industrial revolution in the

late 1700s, the figures were extended to start from 1750 (Steffen et al. 2004). The graphs, which included changes in the human enterprise as well as their impacts on the Earth system, all had similar features: a gradual increase starting with the onset of the Industrial Revolution but then accelerating appreciably around the mid-twentieth century. As evident from Fig. 1, although the precise word “Anthropocene” did not exist at the time, the first pieces of evidence that ultimately led to Crutzen’s concept of this epoch had already appeared in the refereed literature in the 1970s.

The set of Great Acceleration graphs first appeared in the peer-reviewed literature in the 2007 *Ambio* paper “The Anthropocene: Are Humans Now Overwhelming the Great Forces of Nature?” coauthored by Crutzen, Will Steffen, and twentieth-century historian J. R. McNeill (Steffen et al. 2007). It was McNeill, in fact, who first coined the term “Great Acceleration” at an earlier Dahlem Conference workshop in which Crutzen and Steffen also participated (Hibbard et al. 2007). Although the tropospheric ozone trend shown in Fig. 1 did not appear in the original Great Acceleration graphs, trends in atmospheric greenhouse gas concentrations were well-represented in the form of carbon dioxide, methane, and nitrous oxide.

### ***Searching for the missing source of carbon monoxide; hypothesizing “nuclear winter.”***

Based on the calculations in Crutzen and Fishman (1977), Crutzen knew that there had to be a significant source of CO that was not being properly accounted for (e.g., Weinstock 1969). To find the answer to this question, he invited Wolfgang Seiler to Boulder in 1977 to find this missing source. Seiler had studied under Christian Junge in the late 1960s and was generally regarded as the expert on the global CO cycle where he published a comprehensive paper on the global cycle of carbon monoxide (Seiler 1974). One of the initial results of this collaboration between Crutzen and Seiler led to a seminal assessment of the importance of tropical biomass burning on the global budgets of both CO and carbon dioxide (Seiler and Crutzen 1980), suggesting that this source of CO had been grossly underestimated in previous budget estimates. To study this problem, Crutzen organized instrumented aircraft flights to Brazil (using NCAR’s aviation facility) to study the process of biomass burning in more detail (Crutzen et al. 1979).

***Nuclear winter.*** In the fall of 1981 Crutzen received an invitation to contribute an article to a special issue of the journal *Ambio* on the global environmental effects of a major nuclear war. The article was to be part of a larger study sponsored by the Royal Swedish Academy of Sciences that would also include the effects on freshwater supplies, agriculture, ocean and land ecosystems, and medical and psychological consequences for survivors, in addition to the obvious direct effects of the nuclear explosions. The study had been prompted by the heightened tensions between the Soviet Union and NATO countries due to recent deployments of missiles carrying nuclear weapons in both eastern and western Europe. Crutzen invited John Birks, who had just joined him at the Max Planck Institute (MPI) in Mainz for a sabbatical leave from the University of Colorado Boulder, considering that Birks had worked earlier on the effects of past nuclear weapons testing on stratospheric ozone while a graduate student with Harold Johnston at the University of California, Berkeley (Johnston et al. 1973).

The original plan was to update the calculations of stratospheric ozone depletion that had been predicted in earlier studies and confirmed by a report of the National Academy of Sciences (1985) using Crutzen’s two-dimensional atmospheric model. Nitric oxide, recognized initially by Crutzen (1973) to be a catalyst for ozone depletion, is produced from nitrogen and oxygen of air at the high temperatures of the nuclear fireballs, which rise high into the atmosphere. The concentrations and vertical distributions of NO were calculated by Birks for the *Ambio* nuclear war scenario, and Crutzen put those concentrations into his computer model to calculate the results. To their surprise, no depletion resulted. The reason, they soon realized,

was that the *Ambio* scenario of 6,000 total megatons of nuclear explosions was based on the assumption that only modern, smaller nuclear weapons would be used. The fireballs of nuclear explosions having yields less than 1 megaton do not reach the stratosphere. Instead, the NO formed in detonations of a few hundred kilotons is deposited almost entirely in the upper troposphere where oxides of nitrogen ( $\text{NO}_x$ ) have the opposite effect—they catalyze the formation of ozone through the oxidation of CO,  $\text{CH}_4$ , and other hydrocarbons, as shown earlier in Crutzen (1973, 1974). When the scenario was supplemented with extra nuclear explosions of a few megatons each, the Crutzen model did predict the ~50% ozone depletion in the Northern Hemisphere where nearly all of the targets were located and ~20% depletion in the Southern Hemisphere as had been expected.

The discovery that nearly all the NO would be deposited in the troposphere in the small-bomb scenario naturally led to the question of how much ozone and even stronger oxidants such as peroxyacetyl nitrate (PAN) would be formed in the troposphere. Would a “killer” photochemical smog result? Crutzen and Birks realized that the fires ignited by the nuclear explosions would themselves produce additional  $\text{NO}_x$  and, importantly, additional hydrocarbons—another ingredient necessary for ozone formation. As an example, calculations based on previous work by Crutzen on biomass burning (e.g., Seiler and Crutzen 1980), they considered what would happen if the nuclear detonations were to result in the burning of one million square kilometers (i.e.,  $1,000 \text{ km} \times 1,000 \text{ km}$ ) of forest. For this calculation, they dug into the forest fire literature to estimate emissions. Indeed, with these estimated inputs, it was intuitive to think that the model calculations might have resulted in the production of a severe smog—something like the highly toxic situation found in Los Angeles in the 1960s. Subsequently, this pollution would spread throughout the entire Northern Hemisphere. However, there is one more ingredient necessary for in situ ozone formation in the troposphere: and that is sunlight. Most of the fire literature emphasized the quantity of smoke produced in wildfires. It occurred to Crutzen and Birks that their calculations might need to be refined to account for the reduction in solar intensity driving the photochemical reactions (e.g., photolysis of  $\text{NO}_2$ ) that produces ozone. The calculations revealed an unsuspected and arguably the most consequential effect of global nuclear warfare—as much as 99% of solar radiation would be blocked from reaching Earth’s surface due to smoke produced in the nuclear war fires. All life on Earth depends on sunlight, both for warmth and for the food chain, which begins with photosynthesis in plants. The resulting paper, “The Atmosphere after a Nuclear War: Twilight at Noon,” published in *Ambio* in June 1982 (Crutzen and Birks 1982; Birks and Crutzen 1983) and later in book form, was followed 18 months later by a paper by Richard Turco, Brian Toon, Tom Ackerman, Jim Pollock, and Carl Sagan (TTAPS) in the journal *Science* in 1983 that modeled the climate effect for the first time and coined the popular term “nuclear winter” (Turco et al. 1983). These two papers led to a several-year study involving many government agencies and laboratories and evaluation reports by the National Academy of Sciences (1985), the WHO (1987), and the Scientific Committee on Problems in the Environment of the International Council of Scientific Unions (ICSU/SCOPE). The nuclear winter phenomenon has been repeatedly confirmed in updated studies over the years as climate models have continued to improve. In fact, the nuclear winter problem forced climate modelers to include the effects of aerosol scattering and absorption of solar radiation on atmospheric dynamics for the first time, which significantly improved model forecasts for global warming due to the anthropogenic release of greenhouse gases as well. Both Ronald Reagan and Mikhail Gorbachev were aware of the nuclear winter effect, as evidenced by mentions in speeches they made, but it remains uncertain to what extent nuclear winter played in subsequent arms control treaties that have reduced the total number of warheads from more than 50,000 in U.S. and U.S.S.R. stockpiles in 1982 when the Crutzen and Birks paper was published to less than 10,000 held by the United States and Russia today.

## **The societal impact of Crutzen's work**

**Writing for the lay community.** In the late 1980s and early 1990s, perhaps spurred on by the interest of this new phenomenon “nuclear winter,” Crutzen’s work now became part of studies that examined issues that were generally far beyond the focus of the atmospheric sciences community. Crutzen and Tom Graedel, a research scientist at Bell Laboratories at the time, became good friends as a consequence of their attendance at a variety of conferences and other gatherings. Crutzen’s concentration at the time was largely on the chemistry of the stratosphere, while Graedel’s expertise focused on lower-atmospheric gas–particle interactions. On three occasions Crutzen had received invitations to contribute a paper to specialized meetings, and he invited Graedel to coauthor papers on gas–aerosol chemistry with him for those events. Since these collaborations resulted in three detailed papers together and both of them had written many other papers successfully, Graedel approached Crutzen with expanding their ideas and producing a textbook. Eventually, they published *Atmospheric Change: An Earth-System Perspective* in 1993, which was translated into German as *Chemie der Atmosphäre*.

A year later, Graedel and Crutzen received an invitation from the Scientific American Library, which was publishing a series of books for readers with undergraduate degrees in science or engineering who valued modest-length volumes on contemporary topics of interest. They managed to generate a 180-page book titled *Atmosphere, Climate, and Change*, which was subsequently published in German, Dutch, Chinese, and Russian. (To best capture the Dutch audience the Netherlands publisher inverted the order of the authors’ names, which was discovered when the volumes were received.) This volume received the AMS Louis J. Battan Author’s Award for communicating atmospheric science to a broad audience.

**The Vatican connection.** The modern Pontifical Academy of Sciences (PAS) was established by Pope Pius XI in 1936, who gave it its present name although the roots of this organization go back to 1603 when the Accademia dei Lincei (Academy of the Lynxes) was founded as the first exclusively scientific academy by Federico Cesi. One of the first members of that academy of science was Galileo Galilei (appointed in 1610), but the academy did not convene after Cesi died in 1636.

Since its modern establishment in 1936, the PAS has grown increasingly international in character, and the Academy is now a member of the International Council of Scientific Unions (ICSU). The current PAS consists of 80 women and men who have made outstanding contributions in their various fields of scientific endeavor. The Pope approves each appointment after being nominated internally by the body of the academicians. These pontifical academicians participate in study groups and meetings organized by the Academy to examine specific issues (see [www.casinapioiv.va](http://www.casinapioiv.va)).

Crutzen made his first trip to the Vatican in 1983 to take part in a workshop that focused on how chemical events impacted the environment and he presented a talk on how a nuclear war would impact the atmospheric composition. Subsequently, in 1984, the PAS issued an official statement that explicitly warned of the possibility of nuclear winter in the aftermath of a war that used atomic weapons. In 1996, Crutzen was invited to become a PAS academician, and was subsequently elected to the PAS Council in 2001. The Council is an eight-member subset of the PAS that convenes semiannually and makes recommendations to the Vatican as to which topics are to be studied by the PAS. In 2004, Veerabhadran Ramanathan, Crutzen’s close friend and scientific colleague, became an academician and likewise was appointed to the PAS Council in 2011; subsequently, Crutzen and Ramanathan organized an international workshop at the Vatican where the topic was *Fate of Mountain Glaciers in the Anthropocene*. The declaration that emanated from this meeting set the stage for a series of climate meetings at the Vatican that continue to this date, and created an alliance between science, policy, and

religion to address and fight climate change. When Francis became Pope in 2013, one of his major interests was the environment and he stressed the mantra that humankind must take a moral responsibility to care for the planet. Out of this philosophy, the encyclical *Laudato Si'* emerged and was released in time so that the United Nations Framework Convention for Climate Change (UNFCCC) Congress of Parties, meeting later that year in Paris, could refer to it (Fig. 3) when discussing the future of the planet. Out of the Paris climate summit, the Paris Climate Accords were formulated and released.

### **The long-lasting implications of Paul Crutzen's contributions**

***Shaping the vision of the European Space Program's Earth observing capabilities.*** Recognizing the value of collaboration and the sharing of the costs involved in providing access to space for civil research, several European nations came together in the early 1960s to form what eventually became known as the European Space Agency (ESA) in 1975, almost two decades after the creation of NASA. Fortuitously Crutzen had benefited from the embryotic European space effort as a recipient of a fellowship that supported some of his postdoctoral work at Oxford.

Subsequently, one of Crutzen's early research interests was to use the measurements of ozone from satellites to investigate our understanding of the physics and chemistry of the atmosphere. While at NCAR, he worked with Arlen Kreuger, one of his first doctoral students, and Don Heath, both of whom were at NASA's Goddard Space Flight Center (GSFC), to confirm one of Crutzen's predictions that destruction of ozone took place following the deposition



**Fig. 3.** Pope Francis shakes hands with Veerabhadran Ramanathan at the Vatican conference, *Sustainable Humanity, Sustainable Nature: Our Responsibility*, that was organized in May 2014. Information from this conference provided information on scientific and social justice information that would later be used as input for Pope Francis's encyclical *Laudato Si'*. The purpose of this encyclical was to show the Catholic Church's support ahead of the drafting of the Paris Climate Accords, which were to be released in 2015. As a member of the Pontifical Academy's Council, Crutzen was instrumental in persuading the Vatican to take a public stand on the issue of climate change and how it adversely impacted the world's poor. Standing behind and partially hidden by Crutzen is his wife, Terttu. Bishop Chancellor of the Pontifical Academies, Marcello Sanchez Sorondo is seen on the left side of this photo.

of energetic charged particles into the upper atmosphere from a solar proton event (Heath et al. 1977).

Around the same time, Crutzen was also invited to be a member of the Halogen Occultation Experiment (HALOE) Science Team when it was established in 1976 (Russell et al. 1993); HALOE was one of the key instruments on NASA's *Upper Atmosphere Research Satellite (UARS)*, launched in 1991. The suite of instruments on *UARS* was designed to investigate changes in stratospheric O<sub>3</sub>. One focus of *UARS* research was the impact of the release of man-made ozone-depleting species such as chlorofluorocarbons (CFCs), carbon tetrachloride and halons. HALOE and the other *UARS* instruments provided important observations in this respect, helping to confirm the role of CFCs in O<sub>3</sub> destruction, at both middle and polar latitudes.

Directing the Air Chemistry Department at the MPI, one of Crutzen's first innovations was the creation of a research group dedicated to the exploitation of in situ and remote sensing optical instrumentation to measure atmospheric trace constituents. In this group, John Burrows, Dieter Perner, and Wolfgang Schneider began to develop concepts for passive remote sensing of trace atmospheric constituents from space expanding upon the heritage of the NASA GSFC group.

In the science policy area at this time, public concern about the human impact on the atmosphere, especially after the discovery of the ozone hole above Antarctica during austral spring (Farman et al. 1985), led to the German parliament (Bundestag), to create an Enquete Commission, entitled the "Protection of the Earth's Atmosphere." Crutzen was appointed to this commission, which advised and made recommendations on a broad set of issues, which included, in addition to stratospheric ozone depletion, man-made climate change from the release of methane and chlorofluorocarbons and assessing the impact of the release of man-made chemicals on tropospheric composition and air quality. In 1988, the proposal to build Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY), was submitted to ESA as part of its *Envisat* polar-orbiting satellite. *Envisat* was launched in 2002.

A simpler and smaller version of SCIAMACHY, Global Ozone Monitoring Experiment (GOME), was successfully launched in 1995 on a mission of opportunity. As its name implies, GOME's objective was to observe and thereby understand the evolution of global ozone loss, and the beginning of a potential global recovery of stratospheric O<sub>3</sub> as a result of the international actions enacted by the Montreal protocol and its subsequent amendments. In addition, GOME began some very important tropospheric trace gas measurements, including sulfur dioxide (SO<sub>2</sub>) and formaldehyde (HCHO) from tropospheric pollution and revealed the rapid growth of NO<sub>2</sub> and SO<sub>2</sub> resulting from the industrialization of China during the latter part of the twentieth century.

Crutzen's wisdom in promoting remote sensing while at MPI and his continuing scientific support and contributions to the GOME and SCIAMACHY science teams were keys to their success. These pioneering missions have been succeeded by the first operational mission of its type, GOME-2 on the ESA EUMETSAT *MetOp-A*, *MetOp-B*, and *MetOp-C* (2006–28) and the Dutch–Finnish contribution of Ozone Monitoring Instrument (OMI) on NASA's *Aura* satellite (launched in 2004). A new generation of instruments on board the EU Copernicus and ESA Sentinel satellites, is being built and brought into operation (e.g., TROPOMI on *Sentinel-5 Precursor*, operating since 2017). This current generation of instruments improves the spatial and temporal sampling of atmospheric trace gas measurements and will meet the evolving needs of atmospheric science and those of environmental policymakers to assess the changing air quality, the state of atmospheric O<sub>3</sub> and the changing atmospheric amounts and emissions of greenhouse gases and thus anthropogenic climate change.

All these advancements in global observing of the composition of the atmosphere support Crutzen's insight about the synergistic needs for atmospheric measurements in conjunction

with modeling, as a tool to observe and manage the human impact on the Earth system, and thereby to help achieve sustainable development in the evolving Anthropocene.

**Crutzen's contributions put into broader historical context.** There have been many ramifications of Crutzen's introduction of the Anthropocene. As the scientific community began to understand how humankind has impacted the planet, there were calls for "planetary stewardship" (e.g., Steffen et al. 2011). The social science community has contributed to the discourse with several thoughtful and penetrating critiques of the concept (e.g., Malm and Hornborg 2014; Latour 2017; Hamilton 2017; Thomas 2019). Perhaps the most important outcome, however, was the process established in 2009 to formalize the Anthropocene in the geologic time scale (GTS; Head et al. 2023). The process has led to a formal recommendation that the Anthropocene be officially added to the GTS, thus terminating the Holocene (AWG 2019). An active search is underway for the "golden spike," the physical core that would become the marker for the beginning of the Anthropocene (Waters et al. 2016; Head et al. 2021). Thus, Crutzen's Cuernavaca outburst may well herald the beginning of a significant, new time interval in the 4.5-billion-yr history of our home planet.

**In his own words.** Perhaps the best way to summarize the meaningfulness of Crutzen's impact on how we view the planet and how human activities have changed its chemical composition, climate, and even its social fabric, we conclude our tribute by including a passage that he wrote; these words have been taken from the forward he wrote for Al-Delaimy et al. (2020), a collection of papers presented at a Vatican Conference held in 2017:

The notion of the Anthropocene—the age of humans—comes from the observation that humanity is contesting nature in shaping our planet by profoundly changing the atmosphere, biosphere, land surface, and oceans. 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.

For example, the consumption of limited resources, as well as global warming and shifting precipitation patterns, can affect agriculture and jeopardize food security. Global warming and environmental pollution are also challenging issues of justice because the poor and future generations are the ones most affected. Global environmental change and societal disparity can lead to migration, territorial conflicts, and pressure on resources like water and arable land, which in turn can endanger international stability and peace. Thus, the well-being of humans and the integrity of our environment are closely coupled, as expressed by the term "planetary health."

Both the challenges and the opportunities of a safe and prosperous Anthropocene are reflected in Al-Delaimy et al. (2020), including perspectives of sciences, engineering, medicine, humanities, politics, and religions. I hope and wish that the integration of scientific knowledge with societal considerations will help to solve the challenges and grasp the opportunities of the Anthropocene for the well-being of humanity, now and for future generations.

When he died in 2021, the tribute in *Scientific American* stated "Paul Crutzen [may have been] the greatest scientist of *all time*" (Zalasiewicz et al. 2021). Although such a statement is certainly hyperbolic, and the purpose of this article is not to offer a defense to this claim, it does cause one to think why such a statement was ever made in the first place. From the perspective of Earth system science, many well-respected scientists in that field are convinced that the transformation from the Holocene to the Anthropocene, a term clearly defined by



Crutzen in a moment of exasperation, is truly a once-in-a-lifetime event (Zalasiewicz et al. 2016). In summary, we hope that this tribute illustrates why such praise is not entirely out of order.

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**Data availability statement.** No datasets were generated or analyzed during the current study.

### **Appendix: The complete works of Paul Crutzen**

Paul Crutzen’s contributions to science span more than 500 referenceable works and over a dozen books (Crutzen and Brauch 2016); a complete listing is available at [www.mpic.de/4862435/publikationen\\_pcrutzen\\_stand\\_31\\_jan\\_21.pdf](http://www.mpic.de/4862435/publikationen_pcrutzen_stand_31_jan_21.pdf). A comprehensive summary of his scientific work can be found in an article recently published the Royal Society (Müller 2022). This paper does not intend to provide such a broad-ranging summary of his body of work over the half-century it took to compile. Instead, the purpose of this article is to reach out to the atmospheric sciences community and provide a couple of snapshots of Crutzen’s career from a perspective of a few of his colleagues who worked closely with him. In particular, we focus on two broad themes that stand out: the linkage between atmospheric chemistry and the Anthropocene, and Crutzen’s contributions to the lay community that eventually focused on our moral obligation to do as much as possible to understand and mitigate climate change. We acknowledge that Crutzen’s research includes such important areas as geoengineering (e.g., Crutzen 2006) and the relationship between widespread air pollution and climate (e.g., Lelieveld et al. 2001), in addition to several other areas of research have not been mentioned here. On the other hand, the purpose of this paper has been to link several research topics where the linkage is not so obvious. Last, as highlighted in the third part of this paper, we show that his impact will be felt for decades to come, and possibly even for centuries and millennia.

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