

A late change to the programme

How an engineer became hooked on atmospheric chemistry.

Paul J. Crutzen

As a child, I dreamed of being a scientist, but when I left middle school this looked impossible. Because of illness during my final exams, my grades were too low to obtain a stipend to continue to university, and so I switched to a shorter course in civil engineering. I found it interesting — in my first job as an engineer, I helped to construct a pedestrian tunnel between Amsterdam's central railway and the tram stations. Unfortunately, few used it, and so it had a short lifetime. In 1958, I married a Finn and moved to Sweden, where I worked for a construction company. But I had not given up hope of higher learning.

My chance came when the Meteorological Institute of Stockholm University advertised for a computer programmer. There were many applicants and I did not have any experience, but I got the job. So in the summer of 1959, with a baby daughter in tow, my wife and I moved to Stockholm, where I was to learn a new profession — computer programming. At last, at the age of 26, in between writing programs for various meteorological applications, I could attend some lectures at the university. I had to concentrate on theoretical subjects, so I studied mathematics, mathematical statistics and meteorology in my free time.

By the middle of the 1960s I was helping a visiting scientist, James Blankenship, with calculations on the vertical distribution of atomic oxygen and ozone in the upper atmosphere. We were using Sidney Chapman's five 'oxygen-only' chemical reactions, first described in 1930, for how sunlight converts the various forms of oxygen — O , O_2 and O_3 — into one another.

I became hooked on stratospheric ozone chemistry. I avidly began to study the literature, and became fascinated by catalysis, which is so characteristic of atmospheric chemistry. Some studies at that time had suggested that OH and HO_2 radicals control stratospheric ozone. In my *Filosofie Licentiat* thesis (comparable to a PhD) I pointed out that the rate coefficients assumed in those studies — they had not been experimentally measured — could not explain the vertical distribution of ozone and would have led to unrealistically rapid ozone destruction in the troposphere. Soon after, laboratory investigations showed that the reaction coefficients were indeed much too high.



The author (centre) with his *Filosofie Doctor* examiners, Richard Wayne (left) and John Houghton, at Stockholm University.

In 1969, I was awarded a stipend from the European Space Research Organization to join John Houghton's research group at Oxford University as a postdoc. Houghton had developed techniques to measure trace gases in the stratosphere from satellites, and soon after my arrival in Oxford, he showed me optical spectra taken in the stratosphere by David Murcay and colleagues from the University of Denver, Colorado. Intriguingly, these spectra contained features of HNO_3 , I reasoned that if HNO_3 was in the stratosphere, then there must also be NO_2 , as this is produced by photodissociation of HNO_3 by solar ultraviolet radiation.

In 1970, encouraged by these findings, I proposed a simple reaction chain, involving NO and NO_2 as catalysts, that destroys ozone in the stratosphere. Because NO is produced by oxidation of N_2O , which in turn originates from microbial transformations at the Earth's surface, this demonstrated that biological activity strongly influences stratospheric ozone chemistry. In the following year, Harold Johnston and I both called attention to the possibility that the NO emissions from large fleets of high-altitude, supersonic aircraft (that were to be built in the United States, the Soviet Union, Britain and France) would damage the levels of stratospheric ozone. Major international research programmes were started to determine more about the physics and chemistry of the stratosphere — which at that time was also dubbed the 'ignosphere' as so little was known about that

region of the atmosphere. These studies confirmed that at altitudes above roughly 20 km, the NO emissions could indeed deplete ozone levels, but also showed that the original ozone depletion estimates were too high, because the effect of some producing reactions, similar to those that produce ozone in the atmosphere, had been underestimated. Those were nervous years, as I was proposing new atmospheric chemistry without a proper background in chemistry. However, a year spent at Oxford University's Physical Chemistry Laboratory with Richard Wayne helped me immensely.

As it turns out, NO emissions are growing only rather slowly, as the planned fleets of supersonic aircraft did not materialize. The greatest threat to ozone levels was revealed in 1974 by Mario Molina and Sherwood Rowland, who showed that Cl and ClO — photolysis products from industrial chlorofluorocarbons — strongly deplete ozone. In 1985, Joe Farman and colleagues surprised the scientific world when they reported the springtime 'ozone hole' over Antarctica, a place where ozone had always been supposed to be inert. But that is another story.

Despite my late start in academia, I had the great fortune to enter a research field in which there was so much to discover. To describe the chemistry of the atmosphere, we now need hundreds of reactions, many more than Chapman's five reactions. Much knowledge has been gained in atmospheric chemistry since the early 1970s — for example, air pollution does not only come from fossil-fuel burning, but also from extensive tropical biomass burning; and in extreme scenarios, burning cities and industries during a nuclear war could cause 'nuclear winter'. Most research in atmospheric chemistry has to do with humankind's strong interference with nature — we live in the 'anthropocene'. Fortunately, an international agreement has been reached to ban the production of ozone-depleting substances — an important turning point in global environmental policy. Although it is hard to be optimistic, let us hope that agreements on global climate and nuclear disarmament will follow. ■

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