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Rowland F S & Molina M J. Chlorofluoromethanes in the environment. *Rev. Geophys. Space Phys.* 13:1-35, 1975.
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This was the first detailed report on the chemical reactions affecting the chlorofluoromethanes CCl_3F and CCl_2F_2 after their release into the environment. The only important atmospheric removal process for these compounds is photodecomposition in the stratosphere by solar ultraviolet radiation. Such photolysis ultimately results in a long chain reaction that can remove enough stratospheric ozone to become a global environmental problem of major significance. [The *SCI*® indicates that this paper has been cited in over 310 publications.]

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During 1973 I proposed to the US Atomic Energy Commission (AEC) (which had sponsored my radiochemical research since 1956) that we expand our investigations of trace radioactive isotopes in gas-phase laboratory experiments into an examination of the atmospheric fate of the nearly inert chlorofluoromethanes, which two years earlier had been detected in minute quantities in tropospheric air.

The actual investigations began when Mario J. Molina joined my research group as a postdoctoral research associate in 1973. Chlorofluoromethanes are inert under most atmospheric conditions, and we soon deduced that they would survive in the atmosphere for an average of 50 to 150 years before being decomposed by very short wavelength solar ultraviolet (UV) radiation. This UV radiation normally cannot reach the earth's surface because it is intercepted by ozone in the stratosphere above it.

The decomposition of inert chlorofluoromethane compounds releases highly reactive atomic chlorine that proceeds to react with ozone, converting it into ordinary oxygen molecules. When our study disclosed the existence of a chlorine-atom catalyzed chain reaction, the work suddenly took an ominous turn, and we recognized the global environmental problem that became a matter of public knowledge and deep concern during the last half of 1974. (The chlorine-initiated chain reaction was discovered earlier in 1973 by R.S. Stolarski and R.J. Cicerone[1] but had not yet reached the published scientific literature when we discovered it for ourselves.)

Our initial short paper on the chlorofluoromethane-ozone problem was written and submitted to *Nature* in late January.[2] However, the scientific, political, and regulatory ramifications of this research were so widespread that we felt obligated to publish a much more detailed exposition than that contained in the limited space provided for the *Nature* paper. Wary that a journal article of the necessary length would be greatly delayed by the refereeing process, I decided that we should write a detailed report and submit it to the AEC as essentially an unofficial, supplementary report because they had no formal requirement for anything beyond the usual year-end summary of research progress.

Molina and I wrote a long scientific paper during July and August of 1974, and we took five copies of this 150-page AEC report with us to the September 1974 meeting of the American Chemical Society (ACS). These copies proved to be not nearly numerous enough as the chlorofluorocarbon-ozone problem exploded both scientifically and publicly that autumn, and about 250 additional copies of the report were distributed within the next few weeks.

While the paper in *Nature* had passed almost unnoticed, a press conference organized by ACS brought extensive national coverage. The outburst of public interest brought immediate concern at the higher levels of US science policy, and an *ad hoc* panel was hastily convened in late October by the National Academy of Sciences to provide an estimate of the urgency of the problem. I was one of the five members of this panel, which was chaired by Don Hunten. Hunten suggested I publish the AEC report in *Reviews of Geophysics and Space Physics*, and he arranged with the journal's editor, Joe Chamberlain, for its submission as a review paper. Chamberlain and he also decided to consider it as already refereed because Hunten and others had already read it.

This paper was supplanted as the major review of the chlorofluorocarbon-ozone problem only 18 months later by the first reports from panels of the National Academy of Sciences; these have been followed in turn by similar reports every two or three years. The work reported in this article and in its predecessor in *Nature* was the basis for the award of shares of the 1983 Tyler Prize in Environmental Achievement to Molina and me. (Harold S. Johnston received the remaining share of that award.)

The chlorofluorocarbon-ozone problem has again become a topic of pressing scientific interest and public concern. The discovery of massive ozone depletion over the Antarctic in springtime since the late 1970s indicates losses reaching as high as 50 percent in October over a region covering about six million square miles.[3,4] The precise chemical details of this unprecedented ozone loss are still under intensive investigation.[5,6]

The long-term environmental consequences of ozone depletion include an increased incidence of human skin cancer, increased ultraviolet damage to many other biological species, and possible climatic effects achieved through alteration of the temperature structure of the stratosphere. These concerns led to the adoption of an International Convention on the Protection of the Ozone Layer in October of 1985. Agreement was reached in September 1987 on an international protocol for control of chlorofluorocarbon emissions.

1. **Stolarski R S & Cicerone R J.** Stratospheric chlorine: a possible sink for ozone. *Can. J. Chem.* 52:1610-5, 1974. (Cited 170 times.)
2. **Molina M J & Rowland F S.** Stratospheric sink for chlorofluoromethanes—chlorine atomic catalysed destruction of ozone. *Nature* 249:810-2, 1974 (Cited 510 times.)
3. **Farman J C, Gardiner B G & Shanklin J D.** Large losses of total ozone in Antarctica reveal seasonal ClO_x/NO_x interaction. *Nature* 315:207-10, 1985. (Cited 105 times.)
4. **Stolarski R S, Krueger J A, Schoeberl M R, McPeters R D, Newman P A & Alpert J C.** Nimbus 7 satellite measurements of the springtime Antarctic ozone decrease. *Nature* 322:808-11, 1986.
5. **Rowland F S, Sato H, Khwaja H & Elliott S M.** The hydrolysis of chlorine nitrate and its possible atmospheric significance. *J. Phys. Chem.* 90:1985-8, 1986.
6. **Solomon S, Garcia R R, Rowland F S & Wuebbles D J.** On the depletion of Antarctic ozone. *Nature* 321:755-8, 1986.

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