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LETTERS

Ozone Depletion Theory

Gary Taubes' article "The ozone backlash" (News & Comment, 11 June, p. 1580) refers to my commentaries as "purporting to shoot holes in the [chlorofluorocarbon (CFC)] theory of ozone depletion." This is hardly necessary; since March 1988, numerous press releases have announced ozone depletion to be "worse than expected" [from the theory]—thus effectively discrediting it.

My comments have pointed to the lack—so far—of convincing observational evidence for long-term ozone depletion. The data from ground-based observing stations are reported to be contaminated by ultraviolet absorption from atmospheric sulfur dioxide (1); the statistical treatment is inadequate, with the derived "trend" strongly dependent on the time interval selected for analysis (2); and there is also the problem of disentangling any CFC effects from long-term ozone trends of natural origin, correlated with well-recorded trends in sunspot numbers (3).

Obviously, one cannot *exclude* the possibility of a long-term depletion of ozone resulting from anthropogenic causes, specifically CFCs. But with each cause producing its characteristic "fingerprints," proof must rely on a longer time series of more detailed observations (of CFC-specific altitude, latitude, and seasonal dependence).

While skeptical about the evidence for depletion, I consider the Antarctic ozone "hole" to be a genuine phenomenon. I have speculated (4), however, that once there is sufficient chlorine present, the intensity of the hole would mainly be controlled by the presence of polar stratospheric clouds (PSCs)-and therefore by temperature and humidity-rather than just atmospheric CFC concentration. Because the ongoing increase in atmospheric CO₂ should gradually lower stratospheric temperatures (as a result of increased radiation loss) and the increase in methane should gradually increase stratospheric water vapor content (5), it is possible that the hole will persist even if the chlorine concentration falls below the pre-1975 value. We do not know for certain where the chlorine threshold lies: it is possible therefore that an ozone hole could form in the Arctic if climate conditions favor the formation of PSCs there-even in the absence of CFC-produced chlorine (4).

Another controversial issue is covered by Taubes and an adjacent article (F. Sher-

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wood Rowland, 11 June, p. 1571): "What are the relative contributions of natural and human sources to stratospheric chlorine?' One side says the major sources are volcanic (6). The other side criticizes these estimates, arguing that nearly all of the chlorine emitted by volcanic and oceanic sources is washed out in the lower atmosphere, "with negligible quantities reaching the stratosphere" (F. S. Rowland, 11 June, p. 1573). A recent paper (7) postulates a removal of "up to" four orders of magnitude; but Taubes relates that El Chichon increased global stratospheric chlorine by 10%. I conclude that reliable statements about the relative effects of natural and human sources of chlorine should be based on observed trends of stratospheric chlorine rather than on speculative calculations.

Rowland (11 June, p. 1576, reference 13) correctly quotes my views on sources of chlorine as of 1988, but does not cite relevant 1987 papers by Zander *et al.* (8). They found that the total columns of hydrochloric acid (HCl) and hydrofluoric acid (HF) (the major stratospheric reservoir gases for chlorine and fluorine) increased, from 1977 to 1986, at rates of $0.75(\pm 0.2)$ % and $8.5(\pm 1)$ % per year, respectively. Because HF is ascribed entirely to CFCs, the much lower trend for HCl would lead one to believe that there are large natural sources of stratospheric chlorine that overwhelm the CFC contribution.

This situation changed in 1991, however, when Rinsland *et al.*, repeating Zander's measurements of solar infrared spectra, reported increases for HCl and HF of $5.1(\pm 0.7)\%$ and $10.9(\pm 1.1)\%$ per year, respectively, for the period 1977 to 1990, thus suggesting CFCs as a major source (9). Nevertheless, Rinsland *et al.* conclude, and I tend to agree: "In contrast to HF there are significant natural as well as anthropogenic sources of HCl."

According to Taubes, Rowland and others tag their opponents with "selective use of . . . scientific papers and an equally discretionary choice of scientific results" But in his "President's Lecture," Rowland quotes only papers that support his own view on CFC sources; the 1983 paper (10) he cites is in apparent disagreement with Zander's 1987 findings and has been effectively criticized by Prinn (11).

I note in passing that the Montreal Protocol was signed in November 1987, and that production limits on CFCs were tightened in the period 1987 to 1991, when published scientific data indicated that CFCs were not an important source of stratospheric chlorine.

S. Fred Singer

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- R. Zander et al., J. Atmos. Chem. 5, 385 (1987); R. Zander et al., ibid., p. 395. The reported increasing trend for fluorine clearly demonstrates that CFCs are penetrating into the stratosphere; see also the review by F. S. Rowland in Global Climate Change [S. F. Singer, Ed. (Paragon House, New York, 1989)].
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- W. G. Mankin and M. T. Coffey, *ibid.*, 88, 10776 (1983). They report increases of 5% and 12% per year for HCI and HF, respectively, based on aircraft observations between 1978 and 1982.
- 11. R. G. Prinn, in *The Changing Atmosphere*, F. S. Rowland and I. S. A. Isaksen, Eds. (Wiley, New York, 1988), pp. 33–48; Prinn estimates that the results of Mankin and Coffey have a large 1-sigma uncertainty of ±4.5% and ±5.8%, respectively. Thus, the rate of increase of stratospheric chlorine could well be close to zero, in agreement with Zander's 1987 result.

Response: In my presidential address, I referred to Singer's statement that "volcanoes ... contribute substantially to stratospheric chlorine" (1), and I quoted the work of Mankin and Coffey, who showed that the 1982 eruption of the Mexican volcano El Chichon caused only a small increase, less than 10%, in stratospheric hydrogen chloride (2). Singer acknowledges that I quoted him correctly, but objects that I did not cite the relevant 1987 papers by Zander et al. (3). I am pleased to do so now: the data in the Zander papers relevant to a volcanic input of HCl show no evidence whatever for a significant increase in atmospheric HCl during 1982, in agreement with the observations of Mankin and Coffey about the near absence of any influence from El Chichon. There have been only two really large volcanic eruption during the 15 years covered by data on stratospheric HCl-El Chichon in 1982 and Pinatubo in 1991-and neither was an important source of stratospheric HCl.

Prinn indeed estimated that Mankin and Coffey's early data had a large standard error because of the short time span and emphasized that the most efficient method of reducing this error was a longer time series (4). Prinn also discussed the possible negative tropospheric HCl contribution in Zander's data and suggested "that the stratospheric trend may be substantially larger than the Jungfraujoch (that is, Zander's) trend" (4). The continued measurements of Mankin and Coffey had already by 1987 confirmed the reality of their upward trend, which still continues. These additional data were included in the later references to the work of Mankin and Coffey cited in my address (5, 6). The paper by Rinsland et al. in 1991 (7) and another by Zander and colleagues in 1992 (8) confirm that the amounts of HCl and HF found in the stratosphere are fully consistent with carbonbonded molecules as their only significant sources.

The partial sentence of my presidential address quoted directly by Singer as referring to "volcanic and oceanic sources" reads in its entirety, in a paragraph with no mention of volcanoes (p. 1573),

The possible inorganic sources such as hydrogen chloride or sodium chloride from the evaporation of ocean spray dissolve in cloud droplets and are removed by rainfall, with negligible quantities reaching the stratosphere.

However, the work of Tabazadeh and Turco (9) furnishes a detailed scientific rationalization for the absence of a significant stratospheric HCl response to volcanoes, in agreement with the observations of Mankin, Coffey, Zander, and Rinsland *et al.*

The existence of a significant natural source of chlorine for the stratosphere has not been in dispute since the mid-1970s, when the first National Academy of Sciences report stated (10, p. 44): "Methyl chloride . . . evidently has an enormous natural source." However, the contribution of this natural source (about 0.6 parts per billion by volume), which furnished roughly 75% of the stratospheric Cl in 1950, has been easily surpassed by the anthropogenic contributions of CCl₂F₂, CCl₃F, CCl₂FCClF₂, CH₃CCl₃, CCl₄, and so forth, such that CH₃Cl now represents less than 20% of the total Cl input into the stratosphere (11), as cited in my address.

G. M. B. Dobson, the inventor of the spectrometer widely used in ozone measurements, pointed out in 1963 that very large amounts of SO₂ pollution could lead to interpretation as incrementally higher values of ozone (12). De Muer and De Backer have calculated that the ongoing cleanup of massive SO₂ pollution in the vicinity of Brussels requires a correction of 14 Dobson units (out of about 350) in 1972 and only 3 in 1989 (13). They also stated, "It should be clearly noted that we do not want to infer any conclusion about regional ozone trends from this single station analysis" (13). The initial

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levels of SO_2 pollution reported for Brussels are enormous (138 micrograms per cubic meter in 1972), far larger than are now reported for urban pollution in New York or Los Angeles (14), let alone in the locations for most Dobson stations.

Farman *et al.* recognized the importance of possible meteorological changes over Antarctica as potential contributors in their initial report of large observed ozone losses there, but also reported (15)

upper air temperatures, and winds are available for these stations from 1956. There are no indications of recent departures from established mean values sufficient to attribute the changes in total ozone to changes in the circulation.

Solomon et al. (16) then postulated that heterogeneous reactions on polar stratospheric clouds could provide reaction sites for chemical reactions that might result in ozone depletion, noting again that no significant changes in temperature structure prior to the ozone loss had been observed either by Farman et al. (15) or Chubachi (17). The significance of stratospheric temperature changes vis-à-vis Antarctic ozone was then considered by several other research workers in 1986 (18), and the research community quickly concluded that the major effect was that the ozone-depleted Antarctic stratosphere was slower to warm up than in previous years (19, p. 94):

Substantial evidence has been presented for a decline in the temperature of the lower stratosphere over Antarctica since 1979 in October and November.... August and September temperatures show little or no trend over the 1978–1985 period.... This suggests that the ozone depletion has a significant impact on the normal spring radiative warming of the Antarctic stratosphere.

Finally, Singer objects that I quote "only papers that support [my] own view on CFC sources." During the 19 years since the first publications of the connection between chlorofluorocarbons and stratospheric ozone depletion, literally thousands of scientific papers have been published on various aspects of this topic. The ongoing scientific consensus has been expressed in a long series of national and international reports, as cited in my address. Several other comments pertinent to the present discussion follow from the August 1988 NASA report to the U.S. Congress of the results from the international Ozone Trends Panel (19):

The weight of evidence strongly indicates that man-made chlorine species are primarily responsible for the observed decrease in ozone within the [Antarctic] polar vortex (p. 4).

After the solar cycle passed through its minimum in 1985, the impact of solar cycle and trace gases should act in opposite directions on column



ozone... After 1991 when the solar ultraviolet input declines, the total ozone column is again expected to decrease markedly as both effects combine to reduce ozone (p. 72).

The models used in these . . . studies do NOT include the chemistry associated with polar stratospheric clouds that is now believed to be part of the Antarctic ozone "hole". . . . Therefore, neither predictions of the hole, past and future, nor the global or hemispheric significance of Antarctic chemistry, are included in this assessment (p. 103).

The effects of polar stratospheric clouds and heterogeneous chemistry in the Arctic winter [represent major uncertainties] (p. 126).

The measured ozone decreases for these months are not adequately described by an assumed linear variation with time since 1969, but have instead primarily occurred recently rather than gradually over the 17-year period, 1969–1986. An alternate statistical treatment with a nonlinear function whose slope is approximately twice as large in 1986 as in 1965 [that is, proportional to atmospheric chlorine content] provides a set of trend coefficients which is marginally superior to the standard simple assumption of linearity in such trend estimates (p. 37).

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"Critical Fat"

Mike May, in his report in Meeting Briefs (11 June, p. 1592) of a conference I did not attend (I was not invited), made incorrect statements about my research on the relation of menarche and fertility to body fat. The data in my 1974 Science paper (1) are not on runners and swimmers, but on girls and women with weight loss due to excessive dieting. May's statement that normal menstrual cycles "require at least 30% fat by weight (italics mine)" is incorrect. The average for U.S. normal women at completion of growth is about 26% to 28% fat as a percentage of body weight. The minimum or threshold weight for height for menarche represents about 17% fat as a percentage of body

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