

Research News

Arctic Ozone Is Poised for a Fall

An airborne expedition into the Arctic stratosphere found clear evidence that the same cloud-mediated chemistry that leads to the Antarctic ozone hole appears over the Arctic

AS STRONGLY SUGGESTED by ground-based observations made last winter (*Science*, 27 May 1988, p. 1144), the chemical alterations that unleash chlorine to form the Antarctic ozone hole each October are also occurring during the Arctic winter. Preliminary results announced 17 February show that cloud particles formed in the extreme cold of the wintertime Arctic stratosphere convert the chlorine from manmade chemicals such as chlorofluorocarbons into forms capable of destroying ozone.

The Airborne Arctic Stratospheric Expedition was a re-run of the effort that implicated chlorofluorocarbons in the formation of the Antarctic ozone hole. Twenty-three instruments on two research aircraft made 28 flights as far as the North Pole under the coordination of the National Aeronautics and Space Administration (NASA). The expedition could not confirm the actual destruction of ozone, a process that must await strong springtime sunshine in the Arctic stratosphere. Ozone destruction, if it does occur, would not form a hole like that centered on the South Pole; the weather of the northern stratosphere is not right for that. But these results reinforce researchers' suspicions that cloud-mediated ozone destruction is behind the unexpectedly large losses of ozone recently recognized at high Northern Hemisphere latitudes during the winter (*Science*, 25 March 1988, p. 1489).

Expedition members had their fingers crossed when they arrived at their base of operations in Stavanger, Norway, for the 1 January start of flight operations. The Arctic stratosphere is warmer than the Antarctic's, and temperature is the key to the type of ozone-destroying chemicals that they were looking for. A temperature of less than -77°C is required to form nitric acid cloud particles and less than -85°C to form water ice cloud particles. Both types seemed to be central to the release of chlorine from its inactive forms, in which it is tied up as hydrochloric acid or chlorine nitrate, and its appearance as ozone-destroying chlorine monoxide. If this winter had been warmer than normal, as some meteorologists had predicted, there might have been too few polar stratospheric clouds for the high-flying ER-2 or remote-sensor-laden DC-8 to

probe.

The \$10-million expedition might have come up with nothing but a good baseline, but the good luck of stratospheric chemists held. The polar stratospheric clouds began appearing before Christmas, and as of the end of January their abundance was running slightly ahead of the last 10 years' average for an entire winter. The clouds were there and so was the expected perturbation of stratospheric chemistry. Chlorine monoxide reached concentrations as high as 800 parts per trillion by volume (pptv) and were consistently above 400 pptv in sunlit portions of the vortex from 30 January onwards. In the voracious Antarctic hole it was about 1000 pptv. Even outside the swirling vortex of winds that contains the greatest Arctic cold, chlorine monoxide ran from 20 to 100 pptv when models without cloud particle reactions predicted only 0 to 20 pptv.

This much chlorine monoxide was a surprise to researchers. "When we left Norway," says project scientist Adrian Tuck of the Aeronomy Laboratory in Boulder, "we were more concerned than we expected to be. Before the expedition, if you had asked us, we would all have undershot the extent of perturbed chemistry there." As was the case in the Antarctic hole, the increase of the active form of chlorine was reflected in the decrease of inactive reservoirs of chlorine. Concentrations of chlorine nitrate were high at the edge of the vortex and fell toward its interior. The hydrochloric acid reservoir appeared normal outside the vortex but severely depleted inside, at times almost as depleted as that of the Antarctic vortex.

This redistribution of chlorine among its various forms is consistent with laboratory studies of ice-mediated reactions under stratospheric conditions. Reactions on ice surfaces involving the reservoir species release active chlorine and leave behind in the ice the nitrogen that would otherwise rein in the ozone-destroying chlorine. Such ice-mediated reactions happened in the Arctic as well, researchers presume, because abrupt increases in active chlorine monoxide at times coincided with a plane's crossing into an air parcel that had been exposed to polar stratospheric clouds.

Researchers saw some reassuringly familiar chemistry going on in the Arctic, but there were differences that will take some explaining. In the Antarctic, extensive removal of water vapor and nitric acid throughout the lower stratosphere went hand in hand. The formation of water ice helped release active chlorine, and the removal of nitric acid prevented it from being bound up again. Researchers wondered whether denitrification could occur without dehydration. In the Arctic, apparently it can.

On later ER-2 flights, nitrogen in all its forms was reduced at higher altitudes by 90%, while dehydration was limited to a few percent. In contrast, total nitrogen at the lower altitudes of the DC-8 were at times elevated by factors of 3 to 4. That suggests to researchers that nitrogen-laden cloud particles might have fallen from ER-2 altitudes and evaporated at or above DC-8 altitudes. In any case, nitric acid cloud particles, which form at warmer temperatures, seem capable of releasing considerable amounts of active chlorine all by themselves. The extreme cold

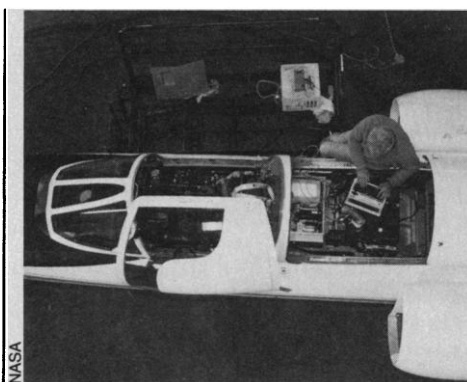


Two ways to sniff out ozone-destroying chlorine. The ER-2 research plane (foreground), a modified version of the U-2 spy plane, flies high with its lone pilot and computerized instruments to make *insitu* measurements while scientists man the DC-8 to make many remote observations.

unique to the Antarctic does not seem required in the Arctic. And even in the warmest years in the Antarctic, which are still colder than the coldest years in the Arctic, there would still be strong ozone-threatening chemical perturbations.

Despite the lower Arctic temperatures and the observed patchiness of both types of polar stratospheric clouds, especially the water ice clouds, the expedition found that "the chemical composition of most of the lower Arctic stratosphere within the vortex has been perturbed." Such widespread alteration probably came about in part by flow-through processing in which a stream of air passes into and out of an area of favorable conditions. The processing zone could be formed as air is forced over mountains, or, as seen on 31 January, by lift provided by a weather system pushing up from below the stratosphere.

"The Arctic atmosphere is primed for a large destruction of ozone," notes Robert Watson, NASA branch chief for its upper atmosphere program and expedition program manager. "The only question is whether the vortex will hold together." If it falls apart before it emerges from its winter darkness, nitrogen from unprocessed air can mix in and squelch the ozone-destroying reactions. If it persists into several weeks of sunlight, sunlight being another essential ingredient in ozone destruction, ozone destruction would be extensive. Unfortunately for anyone wanting to gauge the Arctic's effect on Northern Hemisphere ozone, vortex breakup and the arrival of early spring sunshine usually come within a few weeks of each other in February in the Arctic. (In the Antarctic, the vortex persists many weeks into the sunshine.) When the expedition left Norway on 15 February, sunshine was well into the still intact vortex. Ground-launched, balloon-borne instruments and satellites will follow further developments.



The guts of the matter. A technician works on one of the 13 instruments on the ER-2 aircraft. All are computer controlled.

Still unknown is how much ozone had been destroyed by mid February. The Arctic vortex tends to deform and slide off the pole at times, pushing part of itself into the sun. It is also undoubtedly leaky to some extent. Even the far stronger Antarctic vortex allows some air to pass through it, presumably processing it as it does (*Science*, 12 August 1988, p. 785). The weaker Arctic vortex, which can even break up and reform during the winter, may process a volume of outside air that is a significant fraction of the vortex volume. Expedition flights returned some data that "may be suggestive of ozone loss," according to the preliminary report, but it was too early for extensive losses and further study will be required to say for sure whether any had occurred by then.

Researchers are not making any guesses about the effect of this year's vortex on ozone. They do caution that they are not dealing with another Antarctic hole. The Arctic is simply too different. Once the Arctic vortex breaks up, predicting its effects this year in lower latitudes is not practical either, at least in real time. The analysis and modeling required are just too detailed. The competing and counterbalancing effects of

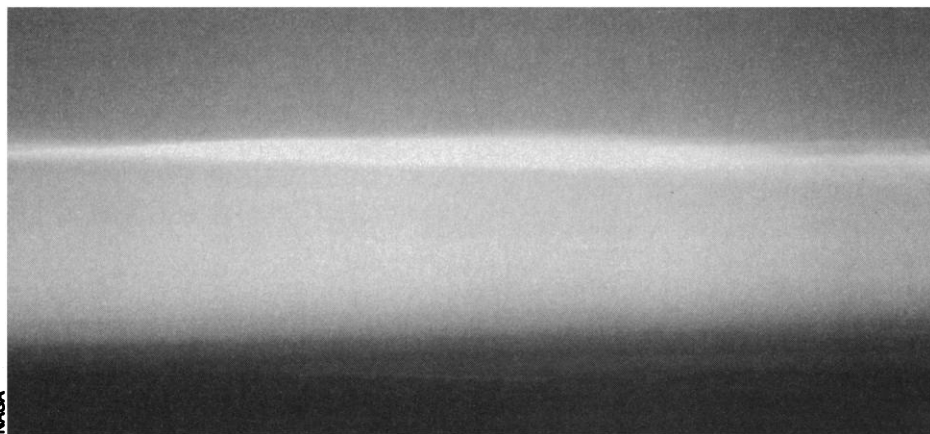
mixing, sunlight exposure, regeneration of residual nitrogen compounds, and further exposure to stratospheric clouds must all be considered. Watson goes only so far as to say that "if the vortex stays together for a few more weeks, there should be an ozone loss."

It might not be too intemperate to guess that the loss would continue the trend of decreasing ozone recently recognized to have been present during recent decades. Last March the international Ozone Trends panel reported that a significant ozone decrease of 1.7% to 3.0% occurred over mid latitudes between 1969 and 1986 (*Science*, 25 March 1988, p. 1489). That fit models predicting losses due to chlorofluorocarbons, but wintertime losses, especially at high latitudes, did not. They ranged as high as 6.2% over the same period. The correlation between ozone loss and cold temperatures, and now the proximity of high concentrations of the ozone-destroying form of chlorine, has reinforced the suspicion that the models failed to predict the losses because they do not include any reactions on particle surfaces.

The inadequacies of the models raise the question of whether the 31-nation Montreal Protocol, which was intended to put a reasonable cap on the destruction of ozone by chlorofluorocarbons, is itself adequate. It was based on the models that lack surface reactions and underestimated the actual ozone loss. Those negotiating the Protocol did have the Antarctic hole glaring at them even as they completed the document and presented it for ratification by its signatories. Thus, not only the models played a role in determining the requirement that chlorofluorocarbon production be cut by 50% by 1999. But, as Watson points out, there have been three new developments since then. Chlorofluorocarbons have been shown to drive the formation of the Antarctic hole, the Northern Hemisphere ozone decrease has been identified, and now the chemistry of the Arctic stratosphere has been shown to be perturbed in a most threatening way.

The scientific review scheduled by the Protocol has already been moved up to August of this year. That will follow by one month a major post-expedition analysis, so that a portion of the results from the mission will be included. A Protocol policy review is then scheduled for 1990. Watson, for one, sees the inevitable doubling of atmospheric chlorine by the middle of the next century allowed under the present Protocol provisions, which would quadruple ozone destruction, as "clearly unacceptable." Further mandated reductions seem inevitable.

■ RICHARD A. KERR



The culprits. The clouds of nitric acid trihydrate near the horizon and water ice clouds banded above play a pivotal role in releasing chlorine in its ozone-destroying form. They form only in polar regions.