The Circuitous Case Connecting Ozone Holes and Space Debris

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Doctors for Disaster Preparedness
20th Annual Meeting
July 27, 2002

Outline:

1. Findeisen and Extraterrestrials – Findeisen proposed that particles produced during the entry of meteorites into the atmosphere stimulate rainfall.

2. Bowen and Extraterrestrials – Bowen sharpened Findeisen’s conjecture by showing a statistical relation between certain visible meteor showers and subsequent days of enhanced global rainfall.

3. Rosinski and Extraterrestrials – In an attempt to test Bowen’s hypothesis, Rosinski discovered a massive influx of extraterrestrial particles apparently unrelated to known meteor showers.

4. Extraterrestrials and the Ozone Hole – Rosinski and Kerrigan showed this influx may provide an alternate explanation for the formation of the ozone hole.

5. Recent Developments – We have progressed in identifying the source of these particles, a mechanism of ozone destruction at particle surfaces, and a method based on these developments to diminish the severity of the ozone hole.

6. A Conjecture – This mechanism may also

   • shape the atmosphere’s temperature profile, radiation budget, and possibly general circulation between 20 and 80 km altitude over the entire globe,

   • preserve our oceans,

   • provide the ice nuclei responsible for Bowen’s days of enhanced rainfall.
Findeisen and Extraterrestrials

A water cloud consists of a large number of microscopic droplets. The droplets are in continual relative motion. When they collide, they tend to bounce off one another. How then does rain form?

The Wegener-Bergeron-Findeisen mechanism (idealized account):

Cloud droplets can be liquid down to about -40°C, and many clouds consist of super-cooled droplets.

An ice crystal in a super-cooled water cloud experiences an environment of relative super-saturation. This effective excess of water vapor condenses on the crystal and promotes rapid growth.

As the crystal grows, its fall velocity increases. At some point, its relative speed in collisions with droplets overcomes the energy barrier to coalescence. The crystal grows even faster by droplet accretion.

Accreted droplets tend to freeze but the freezing is generally incomplete. The transformation of super-cooled water to ice liberates heat, which increases the droplet temperature until it reaches 0°C. At this point, the water and ice coexist in fixed proportions. As the ice crystal grows by accretion, it transforms into a drop of water mixed with ice.

As its size continues to increase, the drop eventually becomes aerodynamically unstable and breaks into four comparably sized fragments (an average). These fragments also grow by accretion of super-cooled droplets and they eventually break up. One drop becomes four; four drops become 16; 16 become 64; 64 become 256; 256 become 1024. One drop becomes about 1000 drops after just five such cycles and 1 billion drops (i.e., rain) after just 15 such cycles.

Special aerosol particles called ice nuclei were thought to be essential in producing ice crystals in clouds and hence precipitation. Yet many more ice crystals were found in clouds than ice nuclei in ground-based measurements. Early in the development of cloud physics, Findeisen proposed that the (potentially) missing ice nuclei were generated by fragmentation of meteorites during entry into the Earth's atmosphere. While still a student, Rosinski began a correspondence with Findeisen in which he took the view that the crystallographic structure of meteoritic particles was unrelated to that of ice and so they would be ineffective as ice nuclei. The debate diminishes at the outbreak of WW II, when the two found themselves on opposite sides of the conflict.
Bowen and Extraterrestrials

E. G. Bowen was one of the three principals in the development of radar, key to England’s victory in the Battle of Britain. He transplanted to Australia and after WW II pioneered radio astronomy and made significant contributions to the art of cloud seeding. Through MI6, he was privy to Findeisen's collected writings. In the mid-50's, he wrote several papers extending Findeisen's conjecture.

Bowen's Hypothesis: Certain visible meteor showers produce particles responsible for worldwide precipitation anomalies, days of markedly enhanced precipitation that follow these showers by 28±2 days.

Rosinski and Extraterrestrials

Rosinski accepted the position of Program Scientist in Aerosol Physics at National Center for Atmospheric Research in the early 60's; I joined his group as a student in 1965.

The specificity of Bowen's Hypothesis permitted experimental testing. Certain Bowen-like days do not have corresponding visible meteor showers. If there are also no particles from invisible showers, then the hypothesis is broken.

To test Bowen's Hypothesis, Rosinski undertook a program of atmospheric sampling at ground stations widely dispersed over the surface of the Earth.

- We sampled 1000 cubic meters of air per day through soluble fiber filters during August, September, and October in 1967, 1969, and 1971.
- We dissolved each filter and extracted iron oxide particles from the solution with a magnet.
- We examined the extracted particles with a scanning electron microscope.

Contrary to expectations, we found large numbers of particles formed into spheres presumably by melting during entry into the atmosphere.
Figure 1: Scanning electron microscope photographs of five magnetic spherules and one spherule fragment. Diameters range from 15 to 20µm. The photograph of the fragment shows that larger spherules may contain cavities.
Extraterrestrial? Absolutely – The graphs show that spherules arrive simultaneously on a daily basis over the surface of the Earth. These correlations in time and space categorically eliminate the possibility of terrestrial origin.

Subsequent chemical and physical analysis showed the spherules share a common source. Associated weakly-magnetic, non-spherical particles appeared to be 100 times more abundant than the spherules themselves.

This discovery asked more questions than it answered:

- The spherules sampled in 1967, 1969, and 1971 are unrelated to Bowen's hypothesis. Extensive aerosol sampling in the 50's established the absence of such spherules, so the validity of Bowen's hypothesis depends on some other particle population.
A mechanism that would initiate annual arrivals over a 10-year time interval eluded us. The evolution of the orbit of comet debris generally occurs much too slowly to account for such rapid onset.

The larger spherules were hollow.

Some cavities contained methane and other carbon compounds, yet carbon was absent from spherule shells.

The crystallography was perplexing: large crystals imbedded in the FeO spherules are supposed to take much longer to form than the fraction of a second for particles to decelerate in the atmosphere.

Particles of various sizes were sampled during relatively short arrival episodes, yet the larger particles settle through the atmosphere in 15 days and the smaller in 90 days. What synchronizes their arrival at ground level?

Chris Junge of the Max Plank Institute at Meinz put the institute's resources at Rosinski's disposal. As a first step, Rosinski sent all particle samples to Max Plank for isotopic analysis. Unfortunately, the samples were lost in a laboratory explosion, and Junge passed away within a few years.

Bowen visited Rosinski at NCAR in ~1973 to thank him for keeping the hypothesis alive in the face of widespread disbelief. Rosinski told me just after the meeting that he didn't have the heart to tell Bowen that he was the one most responsible for killing the hypothesis.

New management at NCAR obstructed further work. Rosinski continues studies in ice nucleation despite nominal retirement and loss of eyesight. I have made my way in the computer industry.

Extraterrestrials and the Ozone Hole

In 1998, before leaving Colorado for Oregon, I stopped in Boulder to say goodbye to Jan. He told me he has just published two papers associating spherule arrivals with the formation of the ozone hole. I thought "What inspired opportunism - finally this data finds a purpose." He asked me to perform just one more computation before disappearing into the woods: Determine whether ozone molecules diffuse to spherule surfaces fast enough to account for ozone loss by oxidation of iron (i.e., rust)?
First thoughts: "The idea is insane, there is not enough iron, the calculation is really short and simple, and the prizes for the consensus chlorine / bromine explanation for ozone hole formation have already been awarded."

Second thoughts: "Working with Jan has always been the best game in town, his intuition is extraordinary, there is always something interesting and important on the verge of discovery, I am incapable of a short or simple computation, and maybe the prizes were awarded prematurely."

So the game was afoot. We labored for two years to try to understand what the data was telling us. Our results are published in two manuscripts:


The object of Part I was to estimate the concentration of extraterrestrial particles in the ozone layer over South Pole, Antarctica, during ozone hole formation.

Observations and Conclusions:

- A major stream of micrometeorites furnishes particles to the ozone layer during formation of the ozone hole.
- This stream is not associated with known meteor showers.
- It has a negative radiant declination and a slow speed of entry into the atmosphere.
- The variability in its intensity from year to year corresponds to the variability in ozone depletion in the ozone hole itself (an exaggeration I regret).
- These particles settle to ground level as aggregates formed in a stratospheric ice crystal coalescence process.
- The concentration of extraterrestrial particles at ozone hole formation lies between 500 and 2000 / m$^3$. 
The object of Part II was to assess the action of particles of extraterrestrial origin in the formation of the ozone hole.

Observations and Conclusions:

- Air in the ozone layer over South Pole tends to be stable, saturated, and essentially particle-free just prior to ozone hole formation.

- Gas-phase reactions generally responsible for ozone loss in the atmosphere are unrelated to ozone hole formation.

- Principal features of ozone hole formation evident in field data for 1986:
  1. the start of ozone hole formation three weeks before sunrise,
  2. a spike up in ozone concentration at sunrise,
  3. cessation in ozone depletion before disintegration of the circumpolar vortex.

![Figure 1.2: Total Ozone Mass vs. Day of Year in a standard column over South Pole, 1986. Sunset is April 3.](image)
A major stream of micrometeorites furnishes particles to the ozone layer during formation of the ozone hole.

Ozone and water vapor diffuse to the surfaces of these particles, where they catalyze a series of reactions leading to net loss of ozone in the atmosphere:

\[
O_3 + O_3 \rightarrow 3O_2, \\
O_3 + H_2O \rightarrow H_2O_2 + O_2, \\
O_3 + H_2O_2 \rightarrow H_2O + 2O_2, \\
H_2O_2 + H_2O_2 \rightarrow 2H_2O + O_2.
\]

These reactions do not require sunlight. \(H_2O_2\) produced before sunrise is decomposed by sunlight. Thus, a fraction of \(O_3\) is regenerated at sunrise (evident in the graphs as the spike).
• Projections of a mathematical model of this system are in good quantitative agreement with the principal features of the field data.

• This analysis has developed considerable evidence that the ozone hole may be a natural phenomenon caused by the depletion of ozone in catalyzed reactions on surfaces of extraterrestrial particles.

The consensus chlorine-bromine theory does not account for the decrease in ozone before sunrise or the spike up in ozone at sunrise; the spherule theory does.

Recent Developments

1. Assess the present state of extraterrestrial particle populations in the atmosphere.

   We hadn't seen spherules in the atmosphere for decades. To determine whether they were still arriving, we partnered with a team at Lincoln High School near Christchurch, NZ, to perform atmospheric sampling from mid-August to mid-October 2001.

   [Image: A pump connected to a fiber filter, with insulation muffling the noise.]
I have included this picture for its quiet dignity. Despite the comic appearance, the design is really well conceived.

Just after we received this all-male lineup, I e-mailed the team that the photo might appear on the cover of National Geographic. Within ten minutes, I received this alternate.
The Lincoln team performed superbly but the outcome was disappointing – the pump failed repeatedly, and the fiber filters were patently insoluble. Nevertheless, we have extracted semi-quantitative evidence that magnetic spherules continue to arrive in abundance on an annual basis.

These photos are typical of spherule findings in both New Zealand and Oregon during ozone hole formation in either hemisphere. Note the cluster of three spherules at the top of the photo on the right.
We hope to sample in New Zealand again this year but with an entirely different technique – no pump, no insoluble filters.

1. Perform a series of laboratory experiments to determine the ability of iron oxide particles to catalyze the destruction of atmospheric ozone.

We have developed a theory in which iron oxide (FeO) is the active agent in catalyzing the destruction of ozone. The essential idea is that iron in an FeO crystal lattice is incompletely oxidized. As a consequence, ozone molecules in contact with the lattice tend to give up oxygen atoms to the particle surface. Incorporation into the lattice is energetically costly and so these atoms may reside at the surface for prolonged periods. Some eventually combine with each other to form O$_2$ or with H$_2$O to form H$_2$O$_2$.

We have partnered with Reinhold Rasmussen on faculty at Oregon Graduate Institute to perform a series of experiments to test this theory. A grant proposal is in preparation.

2. Identify the source of these extraterrestrial particles.

Requirements of any prospective source are severe:

1. The particle stream we sampled decades ago must have recurred every year since but must have also been absent in the decade prior.

2. The stream that provides particles for ozone hole formation over South Pole must also provide particles for ozone hole formation over North Pole six months later.

3. The entire extraterrestrial particle population catalytically active at ozone hole formation must be at least an order of magnitude larger than the sub-population of magnetic spherules used to detect it.

I believe we have found a source that satisfies these requirements. The theory even explains why the spherules are hollow. A manuscript is in preparation.
1. Patent a method to "plug" the ozone hole.

Assuming the chlorine / bromine theory of ozone hole formation, inhibiting formation by direct chemical intervention would require

- a large quantity of reactant to neutralize a large quantity of chlorine, ~60,000 tons,
- solving major reactant dispersal problems,
- engineering around untoward side-effects and unintended consequences,
- identifying a suitable reaction product (probably not a CFC).

Assuming the extraterrestrial particle theory, all these problems are solved by dispersing an agent to modify particle surfaces instead. The mass required, ~9 tons. A patent application is nearing completion.
A Conjecture

The importance of the mechanism we have proposed for the destruction of ozone at South Pole may extend beyond the special environment of the circumpolar vortex. Consider the following possibilities:

The existence of sub-micron particles produced by recondensation in the vapor trails of meteorites was predicted by Rosinski in 1965. Their existence was confirmed by Barbara Kopcewicz about 1975. Frank proposed the injection of water by cometary bombardment of the upper atmosphere (400 tons / min) in 1983. NASA has only recently acknowledged the validity of his concept. Condensation of this water on Rosinski particles finally provides a credible explanation for the production of water-based noctilucent clouds at 80 km altitude.

Ice crystals large enough to make these clouds visible fall rapidly into the stratosphere where they promote the destruction of ozone by the mechanism described in Part II. Modification of the ozone profile may redistribute upper-atmospheric heating, circulation, and transport with possibly important consequences for the Earth’s weather and climate.

The nucleation of ice crystals by Rosinski particles at 80 km and their subsequent gravitational settling also furnishes a mechanism for the net transport of water to lower altitudes. This mechanism not only enables the Earth to capture a fraction of the water made available by small comet bombardment, but also helps to return water of terrestrial origin to lower altitudes, where it is less vulnerable to dissociation by UV and ultimate loss to the planet.

Finally, Rosinski particles that serve as ice nuclei at 80 km are preconditioned to nucleate ice in tropospheric precipitation processes via the Wegener-Bergeron-Findeisen mechanism. This population of ice nuclei may be the connection between meteor showers and Bowen rainfall anomalies we have been looking for for the past 60 years. Thus, we have come full circle.

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