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Materials that enter the stratosphere can remain there for long periods of time, often as stratified layers.
Unperturbed Stratospheric Ozone Distribution
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Shown in the figure below are the results of more recent measurements of \( \text{O}_3 \). The presence of an ozone layer between heights of \( \sim 15-30 \text{ km} \) is very clear.
Mean vertical distributions of ozone concentrations based on measurements at different latitudes (given in degrees).
The $O_3$ layer is **highly variable**. Its height and intensity change with latitude, season, and meteorological conditions. The total ozone column abundance [in Dobson units (DU)] is indicated in the figure.
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Much of the change in the total column abundance of O₃ is due to differences in the profiles below 20 km.
Techniques for Measuring $O_3$

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In this way, a vertical profile of O₃ is obtained, the integration of which provides the O₃ column from ground level up to the height of the balloon.
Ozone Sonde Schematics

Air pump (teflon)  Electrochemical Cell  Transmitter
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This is done by measuring the amount of UV sunlight that reaches the ground, and deducing from this how much UV absorption occurred due to O$_3$. Absorption by O$_3$ occurs in the UV-B region ($\lambda = 0.29 - 0.32 \text{ m}$).
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By taking the ratio of the two measured values, absorption by O$_3$ in the total vertical column can be obtained.
Since 1960 remote sensing measurements from satellites have provided a wealth of information on the global distribution of O$_3$ and the variations in its vertical profiles and column abundance.
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Ozone can be derived from satellite observations using any of the four passive techniques depicted in the following figure:

- backscatter UV (BUV)
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For a description of these methods, see Wallace & Hobbs.
Four passive techniques for measuring ozone from satellites.
Chapman’s Theory
In 1930 Sydney Chapman proposed a simple chemical scheme for maintaining steady state concentrations of O$_3$ in an “oxygen-only” stratosphere. These reactions are called the Chapman reactions.
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- The **dissociation of** $\text{O}_2$ **by** solar UV radiation

$$\text{O}_2 + h\nu \rightarrow 2\text{O}$$
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$$O + O_2 + M \rightarrow O_3 + M$$
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Note that the excess energy acquired by M, which is in the form of thermal energy that warms the stratosphere, derives primarily from the energy of the incoming photon, that is, from solar energy.
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Since the concentration of M decreases with increasing altitude, the time constant for converting O atoms and O$_2$ molecules to O$_3$ increases with altitude (e.g., from a few seconds at 40 km to $\sim$100 s at 50 km).
Schematic illustration of the Chapman reactions.
Stratospheric $O_3$ concentrations exhibit minor diurnal variations (analogous to the day-to-day variations in weather in the troposphere).
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After sunset, both the source and sink of O$_3$ are switched off, the remaining O atoms are then converted to O$_3$ within a minute or so.
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There is an equator-to-pole circulation in the stratosphere, known as the Brewer-Dobson Circulation, which transports O₃ from its primary source in the tropical stratosphere to higher latitudes.
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The destruction of stratospheric O₃ by the various chemical mechanisms discussed above, and many other possible mechanisms not discussed here, are not simply additive because the species in one cycle can react with those in another.

Therefore, advanced numerical models that consider all the known suspects must be used to unravel the relative importance of the various mechanisms for destroying O₃ in the stratosphere.
This task is complicated further by the fact that concentrations of many of the prime suspects are changing with time due to anthropogenic emissions.
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Most of the catalytic reactions that have been proposed for the removal of stratospheric odd oxygen are of the form

\[ X + O_3 \rightarrow XO + O_2 \]
\[ XO + O \rightarrow X + O_2 \]

Net: \[ O + O_3 \rightarrow 2O_2 \]
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If the concentration of catalyst \( X \) is increased significantly by anthropogenic activities, the balance between the sources and sinks of atmospheric \( O_3 \) will be disturbed and stratospheric \( O_3 \) concentrations can be expected to decrease.
One of the first concerns in this respect was a proposal in the 1970s to create a fleet of supersonic aircraft flying in the stratosphere. This is because aircraft engines emit nitric oxide (NO), which can decrease odd oxygen.
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Of much greater concern, with documented impacts, is the catalytic action of chlorine, from industrially manufactured chlorofluorocarbons (CFCs), in depleting stratospheric ozone.
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HFCs and HCFCs are considered to be more “environmentally friendly” than CFCs because they are partially destroyed by OH in the troposphere.
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Such long-lived compounds eventually find their way into the stratosphere where, near an altitude of \( \sim 20 \) km, they absorb UV radiation in the wavelength interval 0.19–0.22 \( \mu \text{m} \) and photodissociate:
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$$\text{CFCl}_3 + h\nu \rightarrow \text{CFCl}_2 + \text{Cl}$$

and

$$\text{CF}_2\text{Cl}_2 + h\nu \rightarrow \text{CF}_2\text{Cl} + \text{Cl}$$
The chlorine atom released by these reactions can serve as the catalyst X and destroy odd oxygen in the cycle

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In 1990, \(\sim 85\%\) of the chlorine in the stratosphere originated from anthropogenic sources. Because CFCs absorb strongly in the infrared, they are also significant greenhouse gases.
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Shown in the next Figure are a series of satellite measurements of the total O$_3$ column in the southern hemisphere.
Satellite observations of the total ozone column (DU) in the southern hemisphere during October for eight years from 1970-2002.
The high O$_3$ values (red and orange colours) that encircle the Antarctic continent and the lower O$_3$ values (green) over the continent itself in the 1970s (upper four images) are considered to be close to natural.
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The Antarctic ozone hole, as it came to be called, is apparent in the lower four images in the Figure as the anomalously low \( \text{O}_3 \) values (blue) over the continent in October from 1999-2002.

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The areal extent of the Antarctic ozone hole, derived from satellite measurements for the period September 7 to October 13 1979-2002 are shown next.
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* * * *

The areal extent of the Antarctic ozone hole, derived from satellite measurements for the period September 7 to October 13 1979-2002 are shown next.

From 1979 to 2001 the ozone hole grew progressively until it occupied an area ($\sim 25$ million km$^2$) similar to that of North America.
Average areal extent of the ozone hole (<220 DU) from September 7-October 13, 1979-2003.
The presence of an ozone hole over the Antarctic raised several intriguing questions.
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Paul Crutzen, Mario Molina, and Sherwood Rowland were awarded the Nobel Prize in Chemistry in 1995 for their explanation of stratospheric O$_3$ loss by CFCs and nitrogen-containing gases.
Mechanism of $\text{O}_3$ Hole
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High-level clouds, called polar stratospheric clouds (PSCs), form in the cold core of the vortex, where temperatures can fall below $-80^\circ$C.

In the austral spring, as temperatures rise, the winds around the vortex weaken, and by November the vortex disappears.
The vortex serves as a giant and relatively isolated chemical reactor in which unique chemistry can occur.
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For example, although the concentrations of O$_3$ in the vortex are normal in August, the concentrations of ClO in the vortex are ten times greater than just outside the vortex collar and, by September, O$_3$ concentrations within the vortex decrease dramatically.
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These decreases are due, respectively, to the formation of nitric acid (HNO$_3$), and to the condensation of water at the very low temperatures inside the vortex.
Polar Stratospheric Clouds
Type I PSCs. These probably consist of a mixture of liquid and solid particles of nitric acid trihydrate $[\text{HNO}_3(\text{H}_2\text{O})_3$ – NAT for short], water and sulfuric acid, which condense at about $-78^\circ$C. These particles are $\sim 1\,\mu\text{m}$ in diameter, so they sediment very slowly ($\sim 10\,\text{m per day}$).
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**Type II PSCs.** These consist of a mixture of ice and water together with some dissolved \(\text{HNO}_3\), form near \(\sim 85^\circ\text{C}\). Since these particles are \(> 10\ \mu\text{m}\) in diameter, they settle out with appreciable speeds \((\sim 1.5\ \text{km per day})\).
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Type III PSCs. These “mother-of-pearl” clouds are produced by the rapid freezing of condensed water in air flow over topography. However, they are of limited extent and duration and do not form over the South Pole.
Schematic of the polar vortex (blue) over Antarctica.
As the particles in PSCs slowly sink, they remove both water and nitrogen compounds from the stratosphere, referred to as **dehydration** and **denitrification**.
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\begin{align*}
\text{ClONO}_2(g) + \text{HCl}(s) & \rightarrow \text{Cl}_2(g) + \text{HNO}_3(s) \\
\text{ClONO}_2(g) + \text{H}_2\text{O}(s) & \rightarrow \text{HOCl}(g) + \text{HNO}_3(s) \\
\text{HOCl}(g) + \text{HCl}(s) & \rightarrow \text{Cl}_2(g) + \text{H}_2\text{O}(s) \\
\text{N}_2\text{O}_5(g) + \text{H}_2\text{O}(s) & \rightarrow 2\text{HNO}_3(s) \\
\text{N}_2\text{O}_5(g) + \text{HCl}(s) & \rightarrow \text{ClNO}_2(g) + \text{HNO}_3(s)
\end{align*}
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[(s) indicates those compounds that are on (or in) ice particles, and (g) indicates species that are released as gases.]
The above reactions convert the reservoir species ClONO$_2$ and HCl into Cl$_2$, HOCl and ClNO$_2$. 
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\text{HOCl} + h\nu \rightarrow \text{OH} + \text{Cl} \\
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Ozone is then destroyed efficiently by

$$\text{ClO} + \text{ClO} + \text{M} \rightarrow (\text{ClO})_2 + \text{M}$$
$$\text{(ClO)}_2 + h\nu \rightarrow \text{Cl} + \text{ClOO}$$
$$\text{ClOO} + \text{M} \rightarrow \text{Cl} + \text{O}_2 + \text{M}$$
$$2\text{Cl} + 2\text{O}_3 \rightarrow 2\text{ClO} + 2\text{O}_2$$

Net: $2\text{O}_3 + h\nu \rightarrow 3\text{O}_2$
Schematic of the polar vortex (blue) over Antarctica. Again!
• It is a **catalytic cycle** in which ClO is the catalyst, because two ClO molecules are regenerated for every two ClO molecules that are destroyed.
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- In the presence of PSCs, Cl$_2$, HOCl and ClNO$_2$ are released and, as soon as the solar radiation reaches sufficient intensity in early spring, Cl and ClO are released, leading to the rapid depletion of O$_3$ in the Antarctic stratosphere.
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The dimer (ClO)₂ is formed only at low temperatures. Low enough temperatures are present in the Antarctic stratosphere before the Spring.
Schematic to illustrate the time evolution of the main processes associated with the development of the Antarctic ozone hole.
(a) The polar vortex.
(b) Chlorine species.
(c) Ozone.
Spring of 2002
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The sharp decrease in the area covered by the Antarctic ozone hole in 2002, and the decrease in the severity of the hole in that year is due to a series of unusual stratospheric warmings that occurred during the winter in the southern hemisphere in 2002.
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In 2003, which was a cold year, the ozone hole returned to its earlier extent of $\sim 25$ million square kilometers.
What about the Arctic?
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However, prior to the winter of 1995–96 there was not a stratospheric ozone hole in the Arctic comparable to that in the Antarctic.
During the northern winter of 1996–97, the longest lasting polar vortex on record developed over the Arctic, and in March 1997 the average ozone column over the Arctic (354 DU) was the lowest in 20 prior years of observations.
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During 1998 a major stratospheric warming event eroded the polar vortex, raised temperatures above that required for the formation of PSCs, and transported ozone into the lower Arctic stratosphere. Consequently, in March 1998 the ozone column over the Arctic returned to its pre-1990s value.
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In the winter of 1999–2000 the average total ozone column in March poleward of 63°N was lower by ~75 DU than the average climatological value, and these low values continued through 2003.
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**Global mean total column ozone** for the period 1997–2001 was $\sim$3% below the 1964–1980 average value.

**No trends** in total column ozone were observed in the tropics ($25^\circ$N–$25^\circ$S) during the period 1980–2000.
Average ozone columns between latitudes $63^\circ - 90^\circ$ for the northern hemisphere in March and the southern hemisphere in October.
Concerns about the health and environmental hazards of increased UV radiation at the Earth’s surface led to international agreements to eliminate the production and use of compounds known to deplete stratospheric O$_3$ by the year 2000.
The International Response

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Consequently, CFCs in the lower atmosphere are no longer increasing. An analysis (in 2003) of 20 years of satellite data showed a slowing in the reduction of O$_3$ at an altitude of $\sim$33 km starting in 1997, and simultaneous slowing in the buildup of harmful Cl.
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Therefore, it is predicted that the O₃ layer will not completely recover until about the middle of the 21ˢᵗ century.
The Montreal Protocol
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Since it takes several decades for all of the air in the troposphere to cycle through the upper stratosphere, where CFCs are broken down, it will take a similar period of time to remove all of the CFCs from the atmosphere from the time their production is brought completely to a halt.
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An Interesting Historical Review

For an interesting review of the emergence of evidence on the ozone hole in the Antarctic, see the article:

Data Collection and the Ozone Hole: Too much of a Good Thing?
On course website.