

Australian wildfires depleted the ozone layer

Various mechanisms initiated by wildfires thinned the stratospheric ozone layer

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The Australian wildfires of late 2019 and early 2020 claimed the lives of 33 people and more than 1 billion animals (1). The fires blanketed southeastern Australia with thick smoke that may have caused the loss of an additional 417 people from hospitalizations associated with elevated levels of fine particulate matter in the air (2). The smoke layer led to a series of atmospheric phenomena that reduced the thickness of Earth's protective ozone layer in the stratosphere, which lies between ~15- and 50-km altitude above the surface. The reduction of total column ozone (TCO), a measure of ozone layer thickness, was particularly strong at mid-latitudes of the Southern Hemisphere during late 2020. Various explanations linking smoke from these wildfires to this observed decline in TCO have been proposed. This debate highlights the shortcomings in our understanding of how wildfires, which are increasingly common because of climate change, affect stratospheric chemistry and Earth's protective ozone layer.

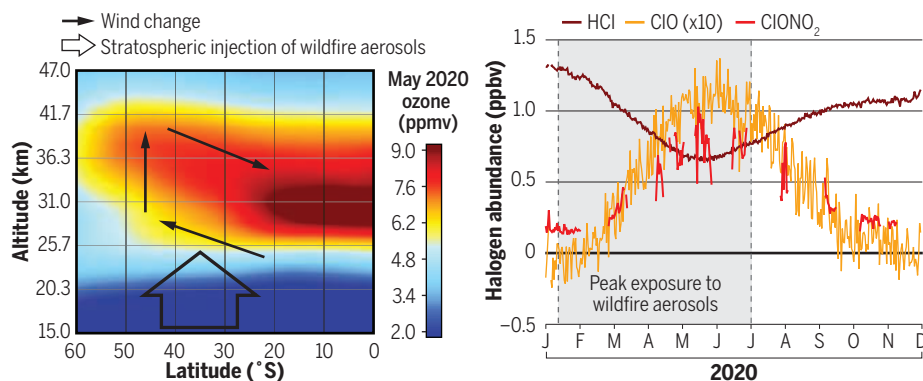
Massive wildfires can initiate or intensify thunderstorms, leading to pyrocumulonimbus (pyroCB) clouds, which are dense, towering structures that often rise to the upper troposphere (12 to 17 km) or lower stratosphere (13 to 22 km) (3). In early 2020, a series of pyroCB clouds connected to the Australian wildfires injected smoke-laden, tropospheric air deep into the stratosphere, reaching an altitude as high as 35 km (4–6). These wildfire-induced convective events are the largest pyroCB clouds ever observed—in terms of both the amount of smoke injected into the stratosphere and the height of the plume—since modern satellite data became available in 1978 (6). The largest smoke-laden plume generated a vortex-like circulation system with a diameter of ~1000 km that persisted for several months and rose to 35-km altitude because of heating driven by the absorption of sunlight by smoke (4–6). As this vortex

moved between 30°S and 55°S, roughly the latitude region of the wildfires, pockets of very low TCO persisted (TCO is the vertical integral of the concentration of ozone from the ground to the top of the atmosphere and is an established measure of ozone layer thickness). The largest reduction in TCO was ~100 Dobson units (DU), meaning that the thickness of the ozone layer was ~60% of the unperturbed value, which is a large decline in the thickness of the ozone layer (5). These reductions in TCO, which existed in ~1000-km-wide structures that traversed the globe every 10 weeks (5), were caused by the injection of ozone-poor tropospheric air into the stratosphere, as confirmed by the associ-

show a substantial increase of SAOD, spanning 30°S to 60°S, that started in mid-January of 2020. The aerosols responsible for the elevated SAOD exhibit a spectral signature characteristic of oxygenated organics (8). The maximum increase of SAOD at Southern Hemisphere mid-latitudes because of the Australian wildfires was larger than the perturbation caused by the April 2015 eruption of the Calbuco volcano in southern Chile yet well below the increase that followed the June 1991 eruption of Mount Pinatubo (9). Both of these eruptions resulted in considerable depletion of stratospheric ozone owing to chemical transformations that were initiated by reactions on the surface of vol-

Response of stratospheric ozone to wildfire aerosols

The thickness of the stratospheric ozone layer in 2020 at Southern Hemisphere mid-latitudes could have been reduced because of changes in wind patterns and by chemical loss reactions involving chlorine monoxide (ClO). The cross section of atmospheric ozone (left) shows changes in stratospheric winds that may have been initiated by the absorption of sunlight by aerosols injected into the lower stratosphere by the Australian wildfires. The chemical process responsible for the observed changes in hydrochloric acid (HCl), chlorine nitrate (ClONO₂), and ClO (right) in the lower stratosphere after exposure of air to wildfire aerosols is important for ozone and is not currently understood.



The latitude/altitude cross section of ozone is from the US NASA Microwave Limb Sounder (MLS) instrument, and the time series show data acquired by the MLS (HCl and daytime minus nighttime ClO) and by the Canadian Space Agency's Atmospheric Chemistry Experiment–Fourier Transform Spectrometer instrument (ClONO₂) (12). ppbv, parts per billion by volume; ppmv, parts per million by volume.

ated simultaneous increase of nitrous oxide (N₂O) and several chemical markers of combustion (4, 6).

As the smoke-laden vortices dispersed, greatly increased concentrations of suspended particulate matter (aerosols) were observed throughout the stratosphere in the Southern Hemisphere (7). The principal method of detecting the aerosols in the stratosphere is to measure the transmission of solar radiation through the stratosphere to the ground, called stratospheric aerosol optical depth (SAOD). Satellite observations

canic aerosols, which predominately have a chemical composition of sulfate (H₂SO₄). The increase of stratospheric aerosols after the Australian wildfires had a number of important consequences for the thickness of the ozone layer: warming of ~1° to 2°C in the mid-latitude lower stratosphere of the Southern Hemisphere (7), the near-immediate alteration of stratospheric winds as a result of this warming (4), and changes in the abundance of reactive chemical species that drive photochemical loss of ozone (7, 10–13) (see the figure).

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Quantification of the effect of wildfire aerosols on ozone is challenging because the concentration of ozone responds to numerous radiative, chemical, and dynamical factors. Furthermore, the nature of surface-mediated reactions occurring on the resulting oxygenated aerosols is uncertain. Soon after the stratospheric injection of smoke by the Australian wildfires had been documented, one study predicted that decreases in TCO of 10 to 20 DU could occur throughout southern middle to high latitudes (40°S to 70°S) owing to changes in stratospheric circulation and chemistry (10). Observations show a substantial reduction in the abundance of ozone in the lower stratosphere between 30°S and 60°S that maximized during May to August 2020 at ~6 to 7 DU below the 2012 to 2019 mean value (7). A different study showed that TCO over much of Southern Hemisphere mid-latitudes during July to November 2020 was ~8 to 15 DU lower than normal, coincident with a substantial reduction in stratospheric nitrogen oxides (NO_x) that are characteristic of the heterogeneous hydrolysis of nitrogen pentoxide (N_2O_5) that occurs on the surface of sulfate aerosols (11). Both studies (7, 11) suggested anomalously low ozone was caused by chemical transformations occurring within the stratosphere, although one study (11) emphasized that wildfire-induced changes in stratospheric wind patterns could also play a role in the reduced TCO observed in 2020.

Some background on stratospheric chemistry helps elucidate the effect of smoke from the Australian wildfires on the ozone layer. The Antarctic ozone hole is caused by transformation of stratospheric inorganic chlorine compounds, most of which originate from anthropogenic activity, owing to heterogeneous (surface-mediated) chemical reactions that occur at appreciable rates only under extremely cold conditions. These heterogeneous reactions cause nearly complete removal of hydrochloric acid (HCl) and chlorine nitrate (ClONO_2), resulting in highly elevated abundances of chlorine monoxide (ClO) that lead to near-complete photochemical loss of ozone. Another heterogeneous chemical reaction, the hydrolysis of N_2O_5 , occurs on the surface of sulfate aerosols for a wide range of temperatures (14). This second heterogeneous reaction converts nitrogen oxides (NO and NO_2) to nitric acid (HNO_3), suppressing the abundance of NO_x and increasing ClO. The net effect of heterogeneous chemical reactions on the surface of sulfate aerosols injected into the stratosphere after recent major volcanic eruptions has been a reduction in the thickness of the ozone layer at mid-latitudes, with little or no variation in the stratospheric concentrations of HCl and ClONO_2 (9).

Chemical processes occurring on the surface of oxygenated organic aerosols injected into the stratosphere after the Australian wildfires led to unusual changes in the chemical composition of the mid-latitude Southern Hemisphere stratosphere that bear some relation to the transformations responsible for the ozone hole (8, 12, 13). In addition to the suppression of NO_x (11), several studies document a large increase in the concentration of ClONO_2 coincident with a substantial reduction in HCl (8, 12, 13). Satellite observations reveal large increases in the concentration of ClO in the lower stratosphere, with a peak in June 2020 that is far above the 2005 to 2019 range of prior observations (12). This increased ClO led to a ratio of reactive chlorine to reservoir chlorine [i.e., $\text{ClO}/(\text{HCl} + \text{ClONO}_2)$] of ~7/100, which is close to that observed after the eruption of Mount Pinatubo (14). However, the SAOD increase after the Australian wildfires was much less than that observed after this eruption. Consequently, the observation of elevated ClO at a level comparable to that of the Mount Pinatubo eruption suggests that the composition of wildfire aerosols is the major determining factor in the observed chemical transformations.

The magnitude of elevated ClO in the mid-latitude Southern Hemisphere lower stratosphere after the Australian wildfires should result in appreciable chemical loss of ozone (9). Nonetheless, several studies attribute the observed reductions in TCO resulting from Australian wildfires to changes in stratospheric wind patterns (12, 13) rather than to chemistry. The evidence for a change in the dynamical state (i.e., wind patterns) of the mid-latitude lower stratosphere of the Southern Hemisphere is strong. Ground-based observations show a rise in the total column (vertical integral) of N_2O , coincident with a decline in total column hydrogen fluoride (HF) starting in May 2020 that is a hallmark of air masses over the vast regions of the Southern Hemisphere becoming less photochemically aged than normal. This meteorological change leads to reductions in TCO because ozone accumulates as air photochemically ages. However, how much of this change in the dynamical state of the Southern Hemisphere stratosphere was caused by atmospheric heating caused by Australian wildfire aerosols is unclear. Before these fires, the direction of winds in the tropical stratosphere had transitioned from a westerly to easterly phase, a phenomenon typically associated with the transport of younger tropical air to mid-latitudes along with an associated reduction in TCO (13).

Understanding the effect of wildfires on stratospheric chemistry is at an early stage. Observations of reduced concentrations of

NO_x and HCl coincident with increased ClO and ClONO_2 are not reproduced by computational models that include known surface-mediated chemical reactions on stratospheric aerosols, assuming that these aerosols have acquired a sulfate coating (12, 13). There is a need for laboratory studies to elucidate the heterogeneous reaction rates that occur on the oxidized organic particles responsible for increased stratospheric aerosol concentrations in 2020 (11). Until such models can accurately simulate observations of ClO after the stratospheric injection of Australian wildfire smoke, it will be difficult to properly assess the degree to which stratospheric photochemistry reduced the thickness of the ozone layer. Increased stratospheric aerosol loading caused by the Australian wildfires may be the driving factor in the severely depleted Antarctic ozone holes of 2020 and 2021 (7, 15) as well as changes in the large-scale circulation of the Southern Hemisphere that extend to the surface (15). The radiative effect of Australian wildfire aerosols on the dynamical state of the Southern Hemisphere should be assessed on the basis of analyses of simulations of stratospheric transport with and without wildfire aerosols, similar to those conducted after the 1991 eruption of Mount Pinatubo (9). Better understanding of the effect of wildfire aerosols on both the chemical and dynamical properties of the stratosphere is needed before consensus is achieved regarding the magnitude of ozone layer depletion attributable to the Australian wildfires. Improved understanding of the threat to the ozone layer posed by massive wildfires is needed because climate change is expected to increase both the frequency and intensity of wildfires (3, 5, 11, 15). ■

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ACKNOWLEDGMENTS

R.J.S. and L.A.M. received financial support from NASA (grant 80NSSC19K0983). R.J.S. and L.A.M. thank S. Varga and B. F. Bennett for helpful input and suggestions and M. Santee and N. Livesey for help with the figure.

10.1126/science.add2056

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Science, 378 (6622), • DOI: 10.1126/science.add2056

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