Atmospheric effects of the Mt Pinatubo eruption

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The eruption of Mt Pinatubo in June 1991 caused the largest perturbation this century to the particulate content of the stratosphere. The radiative influence of the injected particles put an end to several years of globally warm surface temperatures. At the same time, the combined effect of volcanic particles and anthropogenic reactive chlorine has led to record low levels of stratospheric ozone.

It is well known that human activity is perturbing the chemical composition and radiative balance of the Earth's atmosphere. Studies[1] of the sensitivity of our climate to increasing concentrations of greenhouse gases, so named for their ability to retain heat in the atmosphere, predict that the increase of CO_2 concentration from a pre-industrial value of ~270 p.p.m. to 600 p.p.m. by the middle of the next century, along with expected increases in other greenhouse gases, will increase global surface temperature by 2–5 °C. This picture is complicated by the increasing concentrations of anthropogenic aerosols in the lower troposphere, which act to mitigate greenhouse warming[2]. In the stratosphere, chlorine concentrations have increased because of anthropogenic chlorofluorocarbon production[3]. Reactive chlorine compounds play an important role in the chemical processes that give rise to the Antarctic ozone hole, and the weight of evidence suggests that they also contribute to global losses of ozone, with possible concomitant increases in the intensity of biologically harmful ultraviolet-B radiation reaching the Earth's surface[4].

The 15 June 1991 eruption of Mt Pinatubo (15° N, 121° E), on the island of Luzon in the Republic of the Philippines, showed that there is also significant natural, though episodic, variability in the composition of the atmosphere, particularly that of the stratosphere. The Mt Pinatubo eruption forced the evacuation of more than 200,000 people and caused the immediate deaths of more than 300, many from the collapse of homes due to the combination of heavy ashfall and rain from the nearly simultaneous passage of Typhoon Yunya. The threat to life and property from remobilization of tephra ashfall and pyroclastic material persisted through the remainder of 1991 and required careful monitoring by local authorities[5].

The volcanic plume associated with the 15 June eruption was observed to have reached an altitude of more than 30 km. In addition to particulate matter, the eruption also injected gaseous SO_2 into the stratosphere[6], which in turn was transformed into H_2SO_4/H_2O aerosol (particles suspended in air). With an estimated[7] peak aerosol mass loading of 30×10^{12} g (30 Tg), the eruption of Mt Pinatubo caused what is believed to be the largest aerosol perturbation to the stratosphere this century (Table 1, updated from ref. 8), but it is still smaller than the estimated aerosol perturbations from the eruptions of Tambora in 1815 (>100 Tg) and Krakatau in 1883 (~50 Tg)[9]. Given both recent volcanic history and inferences from ancient eruptions such as that of Toba (~1,000 Tg of SO_2) ~75,000 yr ago[9], the 1991 Mt Pinatubo eruption is clearly not exceptional in its modification of the stratospheric aerosol loading. On the other hand, the Mt Pinabuto eruption is unique in the sense that it has been the most intensely observed eruption on record, as its volcanic cloud has been monitored by ground-based and aircraft lidars and solar photometers, various balloon-borne and airborne aerosol counters and other instruments, and many satellite-borne instruments. From a radiative perspective, the aerosol contributed to the end of several years of globally warm surface temperatures experienced in the late 1980s and early 1990s. The aerosol loading has also been associated with chemical and dynamical perturbations affecting the concentration of NO_2, reactive chlorine and ozone. Attempts to model the dispersion of the aerosol and the radiative and chemical impact of the eruption are 'acid tests' of our understanding of atmospheric processes (Fig. 1).

TABLE 1 Major twentieth-century eruptions

Volcano	Date	Estimated aerosol loading (Tg)
Stratospheric background	Possibly 1979	<1
Katmai	June 1912	20
Agung	March 1963	16–30
Fuego	October 1974	3–6
El Chichon	April 1982	12
Mt Pinatubo	**June 1991**	**30**
Cerro Hudson	August 1991	3

Mt Pinatubo aerosol and dynamical processes

Following the 15 June eruption, the evolving cloud of water vapour, sulphurous gases and aerosol moved westwards and circled the globe in approximately 22 days[6]. According to aircraft, ship, and ground-based lidar measurements, the bulk of Mt Pinatubo aerosol was initially concentrated between 20 and 27 km in altitude, although some was observed as low as the tropopause and as high as 30 km. The total mass of SO_2 injected by the eruption was measured[10] by the total ozone mapping spectrometer (TOMS) to be approximately 20 Tg. This SO_2 was eventually converted into H_2SO_4/H_2O aerosol, beginning with an oxidative process involving $OH^{\bullet}$ in a characteristic time[6] of 30 days. The SO_2 mass estimate is consistent with the 30-Tg peak in total stratospheric aerosol mass loading estimated by the solar occultation instrument SAGE II (the stratospheric aerosol and gas experiment-II) in the last few months of 1991.

A notable feature of the Mt Pinatubo eruption was the rapid movement of a substantial fraction of volcanic material across the Equator to about 10° S during the first two-week period following the eruption. Within the next few weeks, the cloud continued to disperse longitudinally, but more slowly in the meridional direction, and occupied the latitude band of approximately 20° S to 30° N (Fig. 2)[11–14]. The degree of early cross-equatorial transport was unusual because the early dispersion of volcanic material from other eruptions near the subtropics, such as El Chichon (17° N, 93° W), remained mostly in their hemisphere of origin. The amount of Mt Pinatubo volcanic material transported south at altitudes above 24 km was highlighted by aerosol observations by SAGE II in July and September and by the improved stratospheric and mesospheric sounder

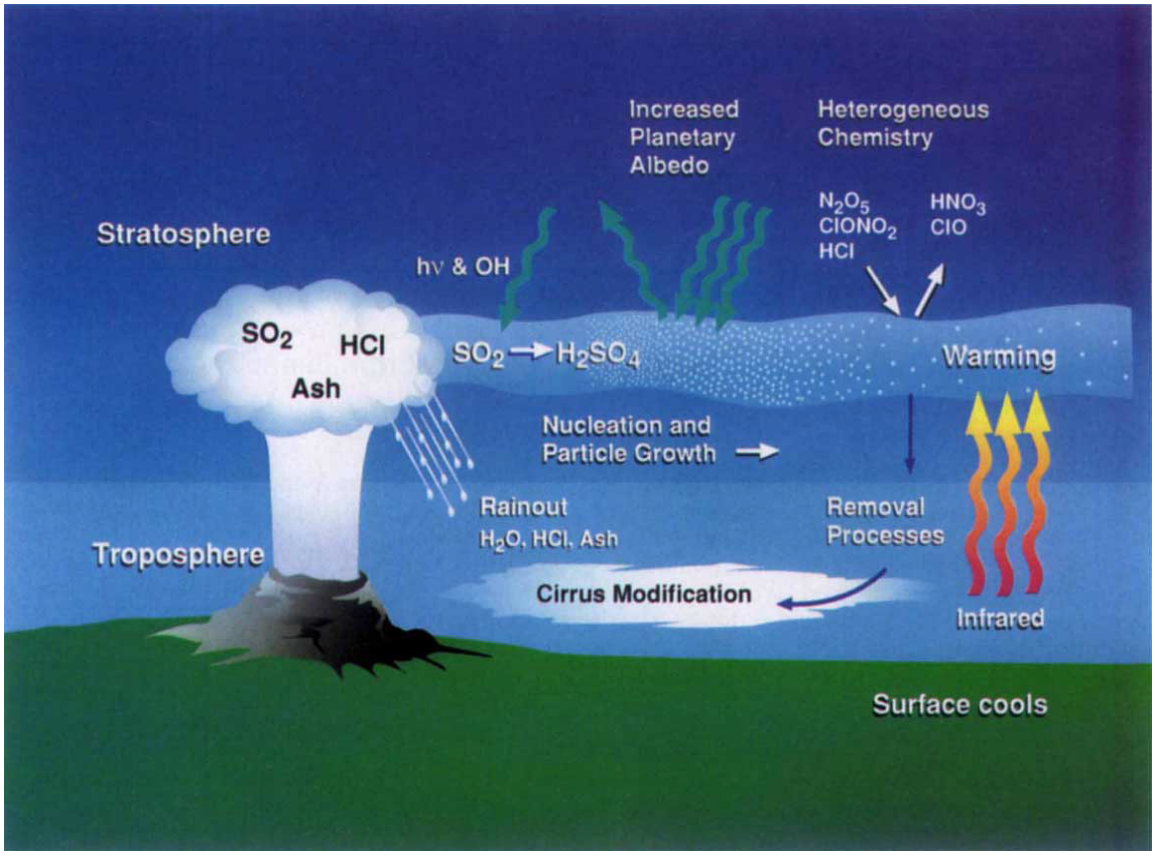

FIG. 1 As shown in this diagram, a volcanic eruption can produce a significant perturbation to the Earth–atmosphere system by injecting material into the stratosphere where, depending on the magnitude and altitude of the injection, it may persist for several years. The injected material may include ash, which typically does not remain for more than a few months, and gaseous components including water vapour, sulphur dioxide and hydrochloric acid. Most hydrochloric acid is dissolved into condensing water vapour and quickly rains out of the original cloud. Aerosols are produced when the sulphur dioxide (SO_2) is chemically transformed into sulphuric acid (H_2SO_4) which rapidly condenses into aerosols because it has a very low saturation vapour pressure. The new aerosol increases the Earth's albedo by reflecting solar radiation back into space, and can warm the stratosphere by absorbing upwelling infrared radiation. Sedimentation and atmospheric circulation eventually transport the aerosol into the troposphere where it may modify cloud optical properties (particularly cirrus) and further modify the Earth's radiative processes. An additional effect of an eruption is the increased efficiency of heterogeneous chemical processes (that is, processes that take place on the surface of aerosols). This effect, coupled with anthropogenically increasing stratospheric chlorine levels, leads to ozone destruction by modifying the chemistry of reactive chlorine and nitrogen. This ozone removal process is similar to that which produces the Antarctic ozone hole, except that the surface in the latter case is provided by polar stratospheric clouds[4].

(ISAMS)[15] in October which showed that maximum aerosol opacity resided near 10° S.

Recent numerical transport simulations suggest that the southward displacement of the aerosol plume within the tropics was the result of meridional circulations induced by local heating, rather than planetary-scale transport patterns[16]. The heating took place through the absorption of upwelling infrared radiation by the newly nucleated aerosol, and was responsible for warming the lower tropical stratosphere through the remainder of 1991. Peak temperatures at the 30-hPa level in the atmosphere (~24 km) were found in September 1991 to be as much as three standard deviations above the 26-year monthly mean (increases of approximately 3.5 K)[17]. The positive temperature anomalies gradually decreased during 1992 as the tropical aerosol dispersed. Strong local heating not only affected horizontal transport, but also enhanced upward motions in the tropics. Satellite observations detected the lifting of aerosol to altitudes exceeding 35 km by October 1991. Because ozone concentration peaks near 25 km in the tropics, a more vigorous circulation at low latitudes, moreover, would have carried ozone-poor air to higher altitudes. At altitudes below 25 km, ozone is long-lived and a gradual photochemical relaxation to pre-eruption levels would have occurred in about 1–3 months. This mechanism could be responsible for a reduction of columnar ozone of about 6–8% observed over the Equator during the first several months after the eruption[18–21].

One feature that seems to be characteristic of large eruptions at low latitudes is the relatively slow transport timescales of volcanic material out of the tropics. Observations following the eruptions of El Chichon in 1982 and Nevada del Ruiz in 1985 revealed the presence of enhanced reservoirs of volcanic aerosol at low latitudes which persisted for several years[22]. Poleward transport of aerosol into the winter hemisphere tends to be suppressed when easterly winds lie over the Equator[23]. During these conditions, strong horizontal wind shear (velocity gradient) lies in the subtropics, separating the tropical easterlies and extratropical westerlies in the winter hemisphere. Wind shear inhibits the horizontal mixing of air. When westerly winds lie over the Equator, however, horizontal wind shear is weaker in the subtropics and meridional mixing occurs more readily. The reversal between easterly and westerly winds in the lower tropical stratosphere is known as quasi-biennial oscillation (QBO). It dominates the circulation in the lower tropical stratosphere and has a period of about 28 months.

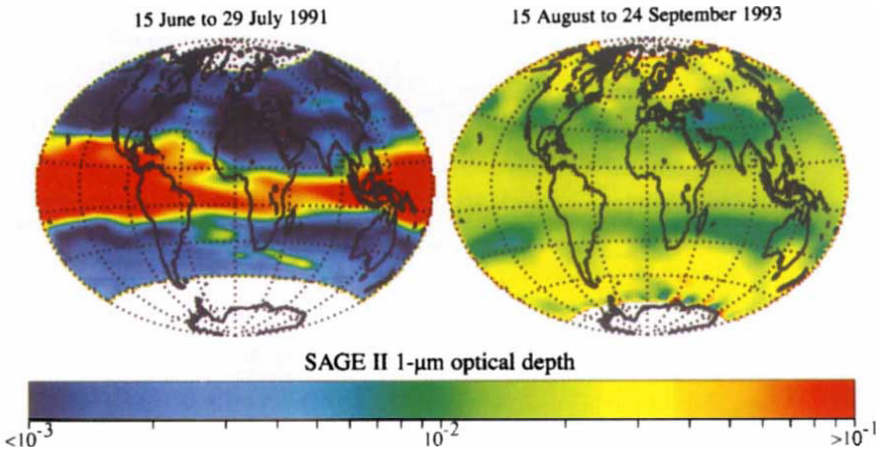

FIG. 2 The development, dispersion, and dissipation of the aerosol from the Mt Pinatubo eruption was monitored by several instruments on satellites. Some interesting observations of the early confinement of the aerosol to the tropical stratosphere were provided by the advanced very high resolution radiometer (AVHRR)[11,14]. Column atmospheric aerosol optical depth determined by AVHRR is usually dominated by boundary-layer aerosols (the lowest few kilometres of the atmosphere) but, after the Mt Pinatubo eruption, possessed a clear volcanic signature until at least early 1993[14]. Several instruments aboard the upper atmosphere research satellite (UARS) were adversely affected by the high concentration of aerosols after the eruption but were also able to provide interesting measurements of the aerosol. For instance, the improved stratospheric and mesospheric sounder (ISAMS) and the halogen occultation experiment (HALOE) made estimates of the relative H_2SO_4–H_2O proportions using strong H_2SO_4 absorption features between 7 and 12 μm (ISAMS) and in the 2.45–5.26 μm wavelength band (HALOE)[15,78]. The stratospheric aerosol and gas experiment (SAGE II), which has been functioning aboard the earth radiation budget satellite (ERBS) since 1984, has made stratospheric aerosol extinction profiles in the visible and near-infrared throughout this period. This figure shows integrated SAGE II stratospheric optical depth for two periods after the eruption of Mt Pinatubo. The data from 15 June to 29 July 1991 (left) show the tropical confinement of the aerosol and the increase (approximately two orders of magnitude) in the 1-μm (actually 1.020-μm) optical depth[7]. Some indications of the initial transport to middle and high latitudes are also evident. SAGE II measurements indicated that by early 1992, the stratospheric optical depth was at, or exceeded, 0.1 at all latitudes. The data for 15 August to 24 September 1993 (right) shows the result of the dispersal and gradual removal of aerosol from the stratosphere. Although the stratospheric optical depth has decreased by approximately one order of magnitude by this time, it is also evident that this optical depth was still dominated by volcanically derived aerosol.

A similar signature of relatively slow tropical aerosol transport to high latitudes was evident after the eruption of Mt Pinatubo. Easterly winds prevailed above about 23 km in the lower tropical stratosphere for the first 6 months after the eruption. Above this height, airborne lidar[24,25] and satellite[26] observations of the volcanic plume found steep meridional gradients of aerosol near 20° N and 20° S. At lower altitudes, however, volcanic aerosol dispersed more quickly towards the poles, and it was primarily this low-altitude transport that was responsible for the initial lidar observations of the Pinatubo aerosol (and the spectacular sunsets) in northern mid-latitudes shortly after the eruption[27–29].

Over the course of time, volcanic aerosol is transported from the stratosphere into the troposphere where a variety of mechanisms (mostly associated with clouds and precipitation) effect the deposition of aerosol to the Earth's surface. Gravitational sedimentation of the aerosol, subsidence (downward air motions) in the winter hemisphere, and episodic transport of stratospheric air into the troposphere through tropopause folds contribute to the removal of aerosol from the stratosphere. By the end of 1993, about $2\frac{1}{2}$ years after the eruption, the global stratospheric aerosol mass loading had decreased to approximately 5 Tg. Since the middle of 1992, the total stratospheric aerosol mass has decreased with a 1/e-folding time of approximately 1 year. Measurements by lidars, Sun photometers, and satellite instruments (for example, SAGE II), showed that the longitudinally averaged stratospheric opacity at a wavelength of 1 μm had declined from a maximum in excess of 0.2 in the tropics in late 1991 to a range over the globe of 0.006–0.03 by mid-1993 (Fig. 2). These values still represent enhancement by a factor of 5–10 of values observed in 1989 and 1990, when the global average stratospheric opacity at 1 μm was between 0.001 and 0.003.

Effect on the Earth's radiative processes

The introduction of large amounts of sulphuric acid aerosol into the stratosphere increases the planetary albedo (essentially the Earth's reflectivity of solar radiation) because these aerosol particles are efficient scatterers but only weak absorbers at solar wavelengths. An analysis of data from the Earth radiation budget experiment (ERBE) aboard the Earth radiation budget satellite (ERBS) found that the albedo in July, August and September 1991 increased compared to a five-year mean[30]. The global albedo in August 1991, for example, was 0.250, or more than five standard deviations greater than the five-year mean of 0.236. The greatest effect occurred over cloud-free regions, where the normally low albedo increased by more than 20%. An increase in albedo was also noted in the normally high-albedo regions associated with deep convective cloud systems. This may represent an indirect effect of the volcanic aerosols in the sense that aerosol particles transported into the upper troposphere are incorporated into deeper convective clouds and alter their microphysical structure[31]. Increasing the number of aerosol particles available to act as cloud condensation and ice nuclei tends to reduce the mean size of the resulting cloud particles. Because smaller particles are more efficient scatterers of visible radiation, this process increases the albedo of the cloud[32]. It has also been suggested that optically thin (but radiatively important) cirrus may be subject to similar microphysical changes[32,33], although it is not at present clear if any changes in the optical properties of cirrus have occurred. The changes in the Earth's albedo observed by ERBE resulted in a net cooling of approximately 8 W m^{-2} between 5° S and 5° N, with a net cooling of 4.3 W m^{-2} between 40° S and 40° N. The meridional distribution of the cooling and its magnitude in the tropics reflects the confinement of the aerosol as discussed earlier.

To a first approximation, an increase in stratospheric opacity, such as that following the eruption of Mt Pinatubo, should cool the Earth because the aerosol increases the albedo of the Earth–atmosphere system and therefore increases the amount of solar radiation scattered back into space. But microphysical properties of the aerosol (in particular, aerosol size) determine whether the infrared absorptivity of stratospheric aerosols, which acts to warm the Earth (that is, the greenhouse effect), dominates the scattering of incoming solar radiation back into space (that is, the albedo effect), which acts to cool the Earth. It has been shown[34] that if the aerosol effective radius (the area-weighted mean radius of the aerosol size distribution) exceeds ~2 μm, the warming effect overrides the cooling effect. Estimates of effective aerosol radius following the Pinatubo eruption indicate that an increase from an average of 0.3 μm before the

eruption to about 1 μm afterwards occurred, and thus cooling would be expected[35–37].

As previously noted, substantial heating in the stratosphere was observed immediately after the eruption. This heating was sufficient to cause tropical stratospheric temperatures to increase as much as three standard deviations above the 26-year mean at 30 hPa (~24 km). Analyses by Christy and Spencer[38] using the satellite-borne microwave sounding unit (MSU) also indicate warmer than average stratospheric temperatures in 1991 and 1992. By the end of 1993, however, stratospheric temperatures had decreased to the lowest values ever observed by MSU. This cooling may be related to the ozone loss observed in the stratosphere (to be discussed later), as ozone is an effective absorber of solar radiation and is responsible for substantial heating in the stratosphere.

The negative radiative forcing associated with the eruption of Mt Pinatubo exceeded the magnitude of the positive forcing associated with the greenhouse gases during the second half of 1991 and much of 1992, and remained significant through 1993 (Fig. 3). In response to this forcing, the mean tropospheric temperature was 0.2 °C below normal in 1992 compared with a base period of 1958–91[39]. The 1992 El Niño–Southern Oscillation (ENSO, a feature of Pacific Ocean circulation with important couplings to tropospheric weather patterns) would have caused a warmer than average troposphere. As a result, the 1992 tropospheric temperature anomaly, adjusted for ENSO[40], was −0.4 °C, a decrease of more than 0.7 °C from 1991. Data from MSU yield a similar global decline in lower tropospheric temperatures of 0.5 °C in mid-1992, with much of the decrease occurring in the Northern Hemisphere (0.7 °C)[41]. In a global-average context, the measured temperature anomaly is consistent with the value of −0.5 °C estimated from a global climate model[42]. It should be noted that the small mean changes in global temperature are not evenly distributed, but rather consist of regions of both relative cooling and warming whose locations and magnitudes are dependent on season[42]. The temperature anomaly in 1993 was smaller than in 1992 with a value of −0.1 °C, which, adjusted for the phase of the ENSO, is equivalent to an anomaly of −0.2 °C (J. Angell, personal communication).

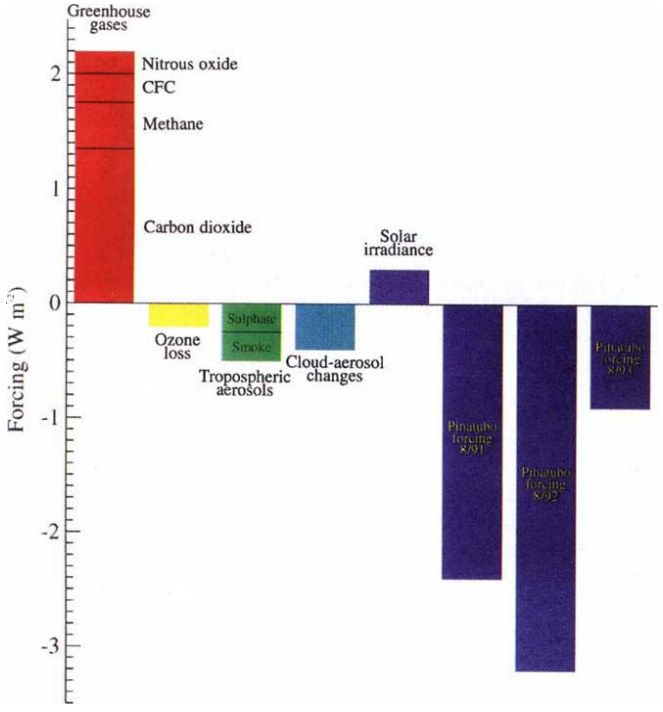

FIG. 3 Pre-industrial-to-modern global climate forcing, 1850–1990 (histogram adapted from Hansen *et al.*[79]). Climate forcing is the change in the Earth's radiative balance resulting from either natural or anthropogenic sources. The largest forcing (~2 W m^{-2}) is due to the increase in greenhouse gases associated with industrialization, and the net forcing in this period remains positive when several smaller (and negative) effects, such as increasing tropospheric opacity due to human activities and global ozone loss, are included. Mt Pinatubo is estimated to have produced a transient negative forcing which exceeded greenhouse-gas forcing between mid-1991 and the end of 1992. It also made 1992 one of the coolest of the past 30 years. As the effect of Mt Pinatubo continues to decrease, the increasing magnitude of greenhouse-gas forcing should reveal itself in a return to the globally warm years observed in much of the 1980s.

This drop in global mean temperatures is similar to that observed in 1964 which had the largest anomaly in the 1958–92 analysis period. Significantly, 1964 is the year following the eruption of Agung, which appears to be the second largest volcanic eruption (after Pinatubo) to have occurred since 1958. The departure is also comparable to the estimated impact of Tambora (−0.4 to −0.7 °C) and greater than that estimated for El Chichon (−0.2 °C) and Krakatau (−0.3 °C)[9]. That the Pinatubo cooling exceeds that of Krakatau is surprising because the latter is believed to have put more SO_2 into the stratosphere. However, this anomaly may simply reflect uncertainties in the estimates of both stratospheric aerosol loading and global surface temperature, particularly for the earlier eruption.

Effect on stratospheric chemical processes

Observations of the input of hydrochloric acid (HCl) soon after the eruption of Mt Pinatubo showed little or no increase compared to pre-eruption levels[43,44]. The measurements are consistent with modelling efforts by Tabazadeh and Turco[45] that suggest that the bulk of erupted chlorine is removed within the volcanic plume by condensation into supercooled water drops which are then rained out. These findings argue strongly that stratospheric chlorine is primarily a product of anthropogenic chlorofluorocarbons rather than volcanic eruptions[46].

The concentrations of chlorine species, and particularly reactive chlorine species such as Cl˙ within the stratosphere are crucial in determining the rate of ozone destruction. Ozone loss associated with reactive chlorine figures prominently in the formation of the Antarctic ozone hole. In the Antarctic, the extreme low temperatures of the winter and early spring permit the formation of polar stratospheric clouds (PSCs)[47]. The cloud particles provide sites for heterogeneous chemical reactions (that occur on the surface of an aerosol or cloud particle) which transform relatively inert forms of chlorine such as HCl and $ClONO_2$ into more reactive forms such as Cl_2. The rates of such reactions are strongly dependent on the total particulate surface-area density. When the chemically modified air is transported into sunlit regions (usually in late August and September at high southern latitudes) Cl_2 disassociates into highly reactive Cl˙ which in turn is responsible for ozone destruction via gas-phase chemical reactions[4].

An additional factor governing the depletion in the ozone hole is that the initial stage of PSC formation involves sequestration of a significant fraction of stratospheric nitric acid (HNO_3) in condensed forms. Because HNO_3 is unreactive, PSCs act as a sink for more reactive forms of nitrogen such as NO_2 and N_2O_5. Reactive nitrogen is a buffer to ozone loss because it is involved in reactions which convert reactive chlorine into non-reactive forms. There is abundant evidence that further growth of PSC particles by the deposition of ice followed by sedimentation of the larger particles leads to an irreversible denitrification (and dehydration) of the polar stratosphere, further enhancing ozone loss[48–50]. The lack of an ozone hole in the Arctic (so far) can be attributed at least in part to generally warmer winter temperatures and, as a result, fewer PSC occurrences[4], less dentrification and a shorter season.

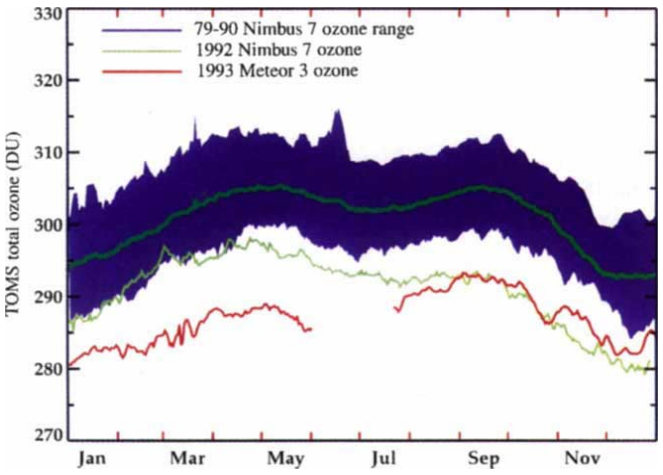

FIG. 4 Global mean ozone in Dobson units (DU) from the total ozone mapping spectrometer (TOMS) measurements as a function of time. Light green line, TOMS/NIMBUS 7 measurements for 1992; red line, TOMS/METEOR 3 measurements for 1993. These two lines are superimposed on the range of TOMS-observed global (65° N–65° S) mean ozone between 1979 and 1990 (dark blue line). The dark green line is the average total ozone for this period. TOMS data show a notable decrease in column ozone amounts beginning in early 1992 and persisting to the end of 1993. The loss reached as much as 6% of the column mean in April 1992[72,73].

After a large eruption such as that of Mt Pinatubo, stratospheric aerosol surface-area density may be large enough for heterogeneous reactions to perturb the concentrations of relevant chemical species not only in polar latitudes but also in middle and tropical latitudes. For instance, ozone loss (although much smaller than that reported below associated with the Mt Pinatubo eruption) attributed to increased aerosol loading was reported following the eruption of El Chichon in 1982[51]. Because elevated chlorine levels in the stratosphere are the result of chlorofluorocarbon emissions, this is an interesting example of the coupling of human and natural perturbations of stratospheric composition that lead to additional perturbations, in this case ozone destruction[52]. It has also been noted that the increase in important greenhouse gases like CO_2, CH_4 and N_2O (as well as CO) have slowed or stopped in the aftermath of the Pinatubo eruption[53]. The cause for these changes is still under investigation.

An early indicator during the post-Pinatubo period of a connection between aerosols and ozone loss was found in association with aerosol formed by the eruption of Cerro Hudson (46° S; August 1991) rather than Mt Pinatubo. Stratospheric aerosol from Cerro Hudson was concentrated at low altitude (<16 km) and was able to penetrate deeply into high southern latitudes soon after that eruption. At that time, Pinatubo aerosol in the Southern Hemisphere was primarily residing above 16 km, with the impedance to meridional transport associated with the polar vortex boundary effectively blocking the aerosol from high latitudes until the vortex dissipated in November 1991[54]. Before 1991, Antarctic ozone loss had not been observed below the base of the vortex (~14 km). But in the densest portions of the Cerro Hudson aerosol layer (11–13 km), ozone decreased by 50% in the 30 days following the arrival of the aerosol over Antarctica[55]. The aerosol surface-area density in the layer was as high as 100 $\mu m^2\,cm^{-3}$, or 20–30 times higher than observed in 1990. Aerosol levels at these altitudes remained unusually high during the austral springs of 1992 and 1993 because of the downward transport of Pinatubo aerosol within the Antarctic vortex[50,56]. Ozone levels in the 12–14 km range were extremely low in 1992 and 1993 with near-zero values in mid-October[57] in both years. Record low column ozone was also observed in these years (for example, 105 Dobson units (DU) on 11 October 1992 and 91 DU on 12 October 1993; column ozone amounts of less than 100 DU were noted on several occasions in 1993[57]).

In the austral autumn of 1992, more evidence of unusual heterogeneous processing was observed over Antarctica. Elevated levels of chlorine dioxide (OClO) were detected over McMurdo station as early as 13 April[58]. Before 1992, elevated OClO was observed only during winter and spring (June–September) as a by-product of heterogeneous chemical reactions on the surface of PSCs. The OClO detected early in 1992 and the ozone losses in 1991 may be the by-products of heterogeneous reactions on the surface of volcanic aerosols:

$$N_2O_5 + H_2O \rightarrow 2HNO_3 \quad (1)$$

$$ClONO_2 + H_2O \rightarrow HNO_3 + HOCl \quad (2)$$

where the moderating influence represented by the photolysis of HNO_3 back to NO_2 (allowing benign $ClONO_2$ to form)[59] was suppressed by the low solar illumination of Antarctica in the autumn. These findings suggest that there was considerable potential for ozone destruction by reactive chlorine in the Antarctic during times when it was too warm for PSCs to occur[58].

On the other hand, evidence of global-scale heterogeneous processing following the eruption of Mt Pinatubo was noted as early as August 1991, when measurements in New Zealand[60] showed a reduction in the column NO_2 that reached as much as 40% in October 1991. Significant column NO_2 decreases have also been reported in northern mid-latitudes[61]. Similar, although smaller, decreases in the column NO_2 were also reported after the eruption of El Chichon in 1982[51,62]. SAGE II measurements showed that the loss of NO_2 occurred globally, largely at altitudes below 25 km and exceeded 50% at some altitudes. Stratospheric NO_2 concentrations remained suppressed throughout 1992 before returning to normal ranges in late 1993. The decreases in NO_2 suggest a repartitioning of reactive nitrogen species in favour of nitric acid (HNO_3) via the heterogeneous reaction (1)[63], a conclusion supported by recent reports of elevated gaseous HNO_3 at low latitudes[64] and mid-latitudes[65–67].

As previously mentioned, column ozone in the tropics decreased by 6–8% in the months following the Pinatubo eruption. The bulk of the loss was observed below 28 km and was as large as 20% near 24–25 km (refs 18, 68, 69). Also, small increases were noted above 30 km. Ozone losses in the lower tropical stratosphere were probably produced by increased lofting associated with stratospheric heating, associated with the new aerosol. In this scenario, ozone is lofted to altitudes at which photochemical processes are more effective in destroying it[19,20]. Other processes may also have contributed to the ozone perturbations in the tropics, including those involving SO_2. Before conversion to sulphuric acid aerosol, SO_2 absorbs ultraviolet radiation thereby reducing the photolysis of O_2 and thus the production of ozone, particularly below 25 km. Above 25 km, photolysis of SO_2 contributes to chemical processes which produce[70] O_3. It has also been suggested that increased scattering of solar radiation by volcanic aerosol may perturb the ozone balance[71].

Six months after the eruption of Mt Pinatubo, global mean column ozone, as measured by TOMS and shown in Fig. 4, began to show a significant downward trend with respect to the mean 1979–90 annual ozone cycle. By mid-1992, the global mean column ozone amount was significantly less than the minimum values measured between 1979 and 1990[72]. This downward trend continued well into 1993; in April of that year a deficit of approximately 6% (~18 DU) was observed, nearly 4% less than ever observed by TOMS before the eruption[72,73]. A slow recovery toward pre-eruption values began in the second half of 1993. It should be noted, however, that global values are somewhat

deceptive because TOMS observed much larger decreases in the Northern Hemisphere than in the Southern Hemisphere[73]. Dobson spectrophotometer observations from several sites in the United States showed a downward trend beginning in May 1992 which accelerated in the first half of 1993, establishing record low values between January and April of that year[74]. On average, these sites measured a decrease of more than 10% compared to the 1979–91 monthly means. Departures were noted throughout 1992 and 1993, although the decrease was smaller in magnitude during May–August. Ozone-sonde data from Boulder, Colorado indicated[75] that this loss was greatest between 13 and 22 km altitude and averaged about 20%. Small increases in ozone concentration were noted above 25 km, though it should be noted that the total column amount is small above that altitude. The altitude of the most significant losses was highly correlated with the altitude of highest aerosol loading.

The observation of highly perturbed values of NO_2 and HNO_3, and the general correlation of the perturbations with enhanced aerosol, strongly indicates the importance of heterogeneous chemical processes in the destruction of ozone. Many of the likely processes are similar to those associated with ozone loss in the Antarctic, in which a loss of reactive nitrogen to non-reactive nitric acid led to enhanced levels of reactive chlorine. The reactions that produce increased reactive chlorine and thus increase ozone destruction include[76] reactions (1) and (2), and other reactions involving $ClONO_2$, HCl and HOCl. Ozone destruction reaches a maximum in the winter and early spring because the direct chlorine reactions are strongly temperature dependent (progressing more rapidly with decreasing temperature), and the photolysis of HNO_3 back to reactive nitrogen (which moderates ozone destruction by reactive chlorine in the lower stratosphere) is inhibited by the low solar illumination. Hanson *et al.*[76] also pointed out that these reactions occur slowly, and that long periods under favourable conditions were required for significant ozone destruction. In the tropics, where solar illumination is high, little ozone loss is noted beyond mid-1992 despite the presence of very high aerosol loading. Modelling efforts by Hanson *et al.*[76] and Rodriguez *et al.*[77] corroborate the importance of heterogeneous reactions in the observed ozone loss.

Looking to the future

The eruption of Mt Pinatubo was a disaster for those living nearby, but for those trying to understand the natural and anthropogenic processes for global change, the eruption represented perhaps a once in a lifetime opportunity. Observations of the Mt Pinatubo aerosol, various chemical species, and the radiative consequences during both the most heavily loaded period and the subsequent recovery provide data needed to test dynamic, chemical and radiative modelling of the atmosphere. Verification of our understanding of the Earth–atmosphere climate system is crucial because future forecasts by such models affect national and international policies and regulations that will in turn directly affect our economic livelihood and quality of life. The Montreal Protocol and its subsequent modifications governing the production of chlorofluorocarbons are good examples of such actions. Of course, much more work is in progress about the effects of Mt Pinatubo than is discussed here. It is also clear that much more work is needed to exploit fully the opportunity provided by nature to enhance our knowledge of how this complex atmosphere and climate system works, and to provide us with a glimpse into the future. □

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