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Volcanoes, PSCs and Ozone Depletion

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It is well known that human activity is perturbing the chemical composition and radiative balance of the Earth's atmosphere. Studies 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 CO₂ 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. In the stratosphere, chlorine concentrations have increased because of anthropogenic chlorofluorocarbon production. 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.

There is also significant natural, though episodic, variability in the composition of the atmosphere, particularly that of the stratosphere. Violent volcanic eruptions 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 (see Figure 1). The injected material may include ash, which typically does not remain for more than a few months, and gaseous components including water vapor, sulfur dioxide, and hydrochloric acid. Most hydrochloric acid is dissolved into condensing water vapor and quickly rains out of the original cloud. Aerosols are produced when the sulfur dioxide (SO₂) is chemically transformed into sulfuric acid (H₂SO₄) which rapidly condenses into aerosols because it has a very low saturation vapor 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 Antarctic ozone hole, except that the surface in the latter case is provided by polar stratospheric clouds (PSCs).

Figure 2 shows a long-term record of stratospheric aerosol column optical depth at a wavelength of 1020 nm obtained from the SAM II instrument aboard the Nimbus 7 spacecraft. The data are weekly-averages for the Arctic and Antarctic regions. The

dates of volcanic eruptions known to have placed material into the stratosphere are annotated on the abscissa. Figure 2 shows that the stratospheric aerosol background levels are dominated by volcanic inputs. The largest in this record is due to the June 15, 1991 eruption of Mt. Pinatubo (15°N, 121°E), on the island of Luzon in the Republic of the Philippines. 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.

The volcanic plume associated with the Pinatubo 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, which in turn was transformed into $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ aerosol with an estimated 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. An extensive discussion of the atmospheric effects of the Pinatubo eruption are described in a recent review article in *Nature Magazine* by McCormick, Thomason and Trepte, Volume 373, 2 February 1995, pages 399-404, which forms the basis for this paper.

In addition to the changes in stratospheric optical depth caused by volcanic eruptions, figure 2 shows seasonal enhancements each winter in both polar regions due to the formation of PSCs. In the Antarctic, the extreme low temperatures of the winter and early spring permit the formation of PSCs. The cloud particles provide sites for heterogeneous chemical reactions which transform relatively inert forms of chlorine such as HCl and ClONO₂ into more reactive forms such as Cl₂. 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) Cl₂ disassociates into highly reactive Cl, which in turn is responsible for ozone destruction via gas-phase chemical reactions.

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 (HNO₃) in condensed forms. Because HNO₃ is unreactive, PSCs act as a sink for more reactive forms of nitrogen such as NO₂ and N₂O₅. 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. The absence to date of an ozone hole in the Arctic can be attributed at least in part to generally warmer winter temperatures and, as a result, fewer PSC occurrences, less denitrification, and a shorter cloud formation season.

Closing Comments

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 modeling 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. It is clear that much more work is needed to exploit fully this 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. Certainly, programs like NASA's Mission to Planet Earth with its earth-orbiting satellites are crucial to understanding global change.

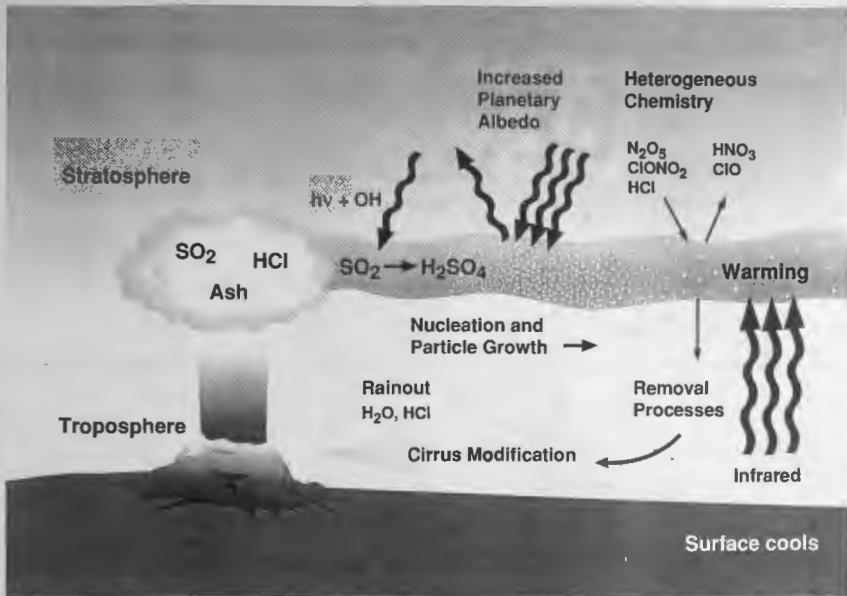


Figure 1. Global effects of violent volcanic eruption

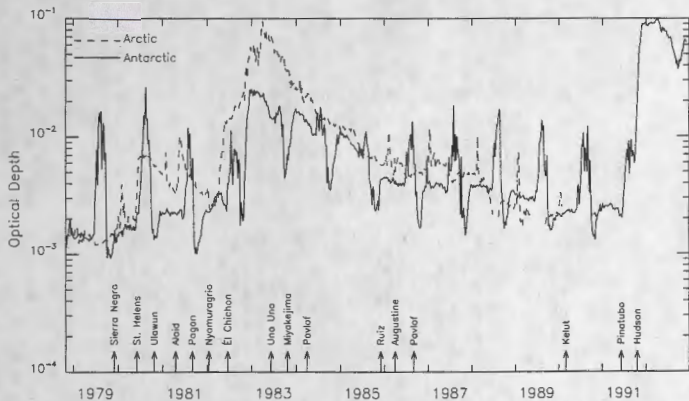


Figure 2. Stratospheric column optical depth at $\lambda=1020$ nm obtained from the Stratospheric Measurement-II (SAM II) instrument.