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## PSC AND VOLCANIC AEROSOL ROUTINE OBSERVATIONS IN ANTARCTICA BY UV-VISIBLE GROUND-BASED SPECTROMETRY

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### ABSTRACT

PSC and stratospheric aerosol can be observed by ground-based UV-visible spectrometry by looking at the variation of the color of the sky during twilight. A radiative transfer model shows that reddenings are caused by high altitude (22-28 km) thin layers of scatterers, while low altitude (12-20 km) thick ones result in blueings. The color index method applied on 4 years of observations at Dumont d'Urville (67° S), from 1988 to 1991, shows that probably because the station is located at the edge of the vortex, dense PSC are uncommon. More unexpected is the existence of a systematic seasonal variation of the color of the twilight sky -bluer at spring- which reveals the formation of a dense scattering layer at or just above the tropopause at the end of the winter. Large scattering layers are reported above the station in 1991, first in August around 12-14 km, later in September at 22-24 km. They are attributed to volcanic aerosol from Mt Hudson and Mt Pinatubo respectively, which erupted in 1991. Inspection of the data shows that the lowest entered rapidly into the polar vortex but not the highest which remained outside, demonstrating that the vortex was isolated at 22-26 km.

### 1. INTRODUCTION

Polar Stratospheric Clouds (PSCs) and volcanic aerosol are involved in polar ozone depletion because of the heterogeneous chemical reactions which take place on the surface of their particles and therefore deserve to be monitored. A method was proposed for PSC routine detection from the ground by UV-visible spectrometry of the zenith sky at twilight. Its validity was demonstrated from a single winter observations in northern Scandinavia, to be operative under all weather (Sarkissian et al., 1990). The method involves monitoring the variation of a sky color index between 90° and 93° solar zenith angle (SZA) at twilight, which is observed to stay constant in clear stratospheric condition, to shift to the red in presence of high altitude (22-28 km) geometrically thin PSC, and toward the blue in presence of low altitude (12-20 km) thick ones. Four years of data are now available at Dumont d'Urville (66°S, 140°E) in Antarctica, which allow the investigation of the seasonal evolution of appearance of PSCs above the station and in addition, in 1991, to trace the arrival of volcanic aerosols injected into the stratosphere by the eruption of Mt Pinatubo in Philippine Islands in June and Mt Hudson in Chile in August.

### 2. INSTRUMENT

Spectral observations are performed with a broad-band (300-600 nm) and low resolution (0.6 nm), diode array UV-visible spectrometer SAOZ (Pommereau and Goutail, 1988). Designed for measuring total ozone and NO<sub>2</sub> twice a day at twilight, the instrument makes observations of the sunlight scattered at zenith by steps of 300 seconds during daytime until loss of sensitivity, that is about 94-95 °SZA, depending of the cloud cover. Data analysis, conducted in real time, allows to calculate by a differential method the molecular absorptions by O<sub>3</sub>, NO<sub>2</sub>, O<sub>4</sub> (collision induced absorption bands of O<sub>2</sub>) and H<sub>2</sub>O. The corresponding absorption features are then removed, leaving at the end of the process a spectrum whose characteristics (and therefore the color), depend on the scattering properties of the atmosphere by molecules (Rayleigh) and particles (Mie) only.

### 3. COLOR INDEX METHOD

In order to characterize the color of the sky at zenith, a color index (CI) was defined as the ratio between fluxes observed simultaneously at 550 and 350 nm, corrected for molecular absorptions. Uncertainties in CI originate mostly from the removal of absorption by ozone in the Chappuis bands with a precision of the order of 1% (Goutail et al., same proceedings). The CI, sensitive to scattering at all altitudes including the troposphere, varies from one day to another, from the blue during clear days to the red in cloudy conditions, the largest CI (red) being observed in extreme scattering conditions during fog or blizzard episodes. In general, during twilight and whatever is its absolute value, the CI is observed to stay almost constant between 90° and 93°, except sometimes in winter, when large variations can occur. Because the troposphere is already in the darkness at twilight, these large changes must originate in the stratosphere. They are caused by PSCs. It is then possible to define a CI variation index or  $\Delta CI = CI(SZA) / CI(90^\circ)$ , whose properties during twilight will depend of the scattering properties of the stratosphere only, providing the tropospheric cloud cover does not vary too much during the one hour twilight period.

A simple scattering radiative transfer model was used to simulate the color index variation (Sarkissian et al., 1990). The atmosphere is divided into 1 km thick spherical shells in which absorption and scattering are calculated. According to the model, reddenings ( $\Delta CI > 1$ ) result from high altitude (22-28 km) and geometrically thin (1-5 km) scatterers. The amplitude at 92-93° SZA increases with the altitude and optical

thickness of the cloud and decreases with its geometrical thickness. Blueings ( $\Delta CI < 1$ ) are generated by low altitude (12-20 km), geometrically thick (5-10 km) scatterers. The blueing amplitude increases with the optical and geometrical thicknesses of the layer.

#### 4. FOUR YEARS OF OBSERVATIONS AT DUMONT D'URVILLE IN ANTARCTICA

Data recorded from January 1988 until the end of 1991 at Dumont d'Urville are shown in figure 1: CI at 90° SZA (upper panel),  $\Delta CI$  between 90 and 92.5° SZA (middle panel) and 50 hPa temperature measured by radio-sondes (lower panel). CI and  $\Delta CI$  reported twice a day at twilight are shown together with 30 days running means.  $\Delta CI$  data are missing in summer during the polar day when the sun zenith angle does not reach 92.5° at midnight and in August 1990 (missing data). Temperature data are often missing in winter because of balloon bursts at low temperature. The problem was partly overcome after August 1990 by the use of an oil dipping method for protecting the balloon.

Day to day and seasonal variations of CI and  $\Delta CI$  appear very similar from one year to another until August 1991, where the first volcanic aerosol arrived above the station making the color indices to depart significantly from the previous picture. The two periods will be discussed separately.

##### Before volcanic eruptions

Daily fluctuations of CI at 90° SZA are generally of about 20%. They result from weather changes. Large reddening occur from time to time during blizzard or snow showers episodes, which result in large enhancements of multiple scattering.

Daily fluctuations of  $\Delta CI$  sensitive to stratospheric scattering only as said above, are generally small (15%) during the warm stratosphere season and before August 1991. Spikes in  $\Delta CI$  are not correlated with spikes in CI demonstrating that the tropospheric contribution is efficiently removed in the  $\Delta CI$ . The relation between extreme  $\Delta CI$  and

PSCs is demonstrated by the plot shown in figure 2. Large  $\Delta CI$  departures occur at temperature below 193 K only, that is the one of formation of PSCs. Year 1988 was particularly rich in events compared to the following years. PSCs are present generally from May until the end of September. This is consistent with SAMS II observations above Antarctica (McCormick et al., 1989). Because Dumont d'Urville is located at the edge of the vortex, stratospheric temperature are not very cold and dense PSCs giving rise to extreme  $\Delta CI$  are unfrequent compared to what was observed above northern Scandinavia in January 1990 (Sarkissian et al., 1990).

Figure 3 shows the observed evolution of  $\Delta CI$  during twilight during the 3.5 year compared to model calculation without volcanic aerosol in the stratosphere, but including background aerosol. Except for few cases when large reddening and blueings are observed in presence of dense PSCs, the model is consistent with the observations.

More unexpected, and which is responsible for most of the dispersion in figures 2 and 3, are the systematic CI and  $\Delta CI$  seasonal variations by some 20%: the twilight sky is bluer at spring than in autumn. To simulate this, the model requires an enhancement of scattering ( $\tau = 0.1$ ) between 8 and 12 km at spring. Two possible interpretations are proposed:

a) the formation of cirrus clouds at or above the tropopause because of the seasonal temperature minimum between 8 and 12 km in August and September, delayed compared to all other levels below and above (figure 4).

b) a large downward diabatic transport of the background aerosols inside the vortex in winter and/or sedimentation by PSC crystals condensed onto the particles, as proposed by McCormick and Trepte, (1987) and McCormick et al., (1989). However, this does not seem to be enough. Relatively blue CI and  $\Delta CI$  are still present at spring after the vortex has moved away from the station, and the model shows that even if all background stratospheric aerosols were transported downward between 10 and 14 km, the result will not exceed a blueing by 5%. A larger optical thickness is required.

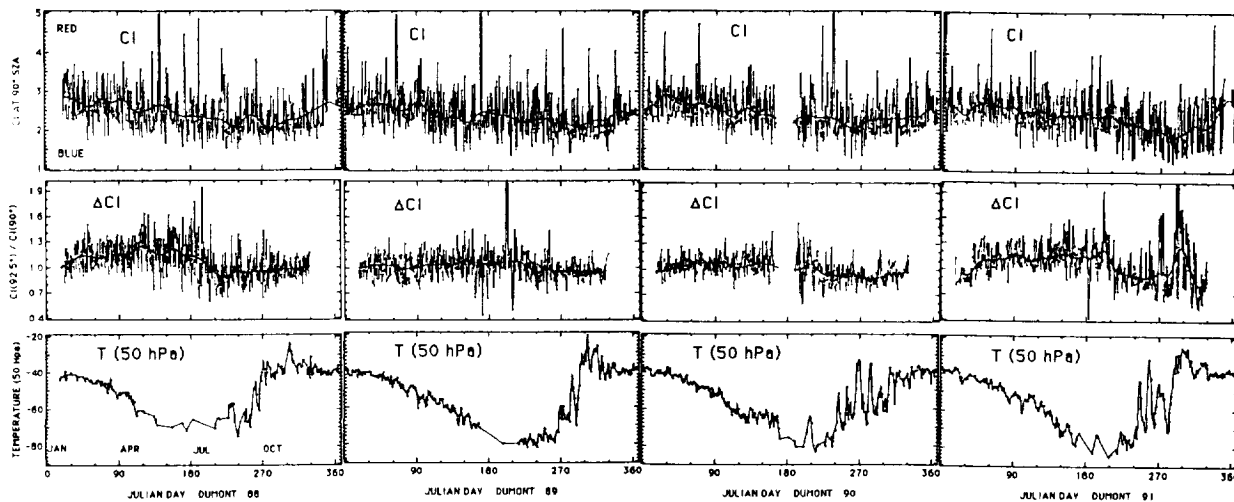


Figure 1. Color index, CI, at 90° SZA (upper panel), color index variation,  $\Delta CI$ , between 90° and 92.5° SZA (middle panel) and 50 hPa temperature observed at Dumont d'Urville from 1988 to 1991.

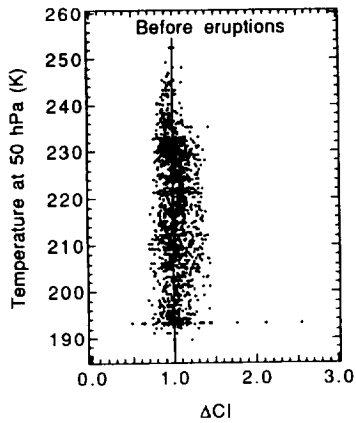


Figure 2. Relation between  $\Delta CI$  and 50 hPa temperature. Large departures are observed to occur at temperature below 193 K only. They are caused by PSCs.

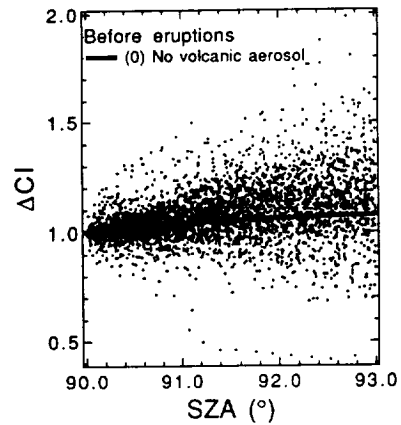


Figure 3. Color index variation at twilight ( $\Delta CI$ ) observed during 3.5 years, and calculated with a simple scattering model.

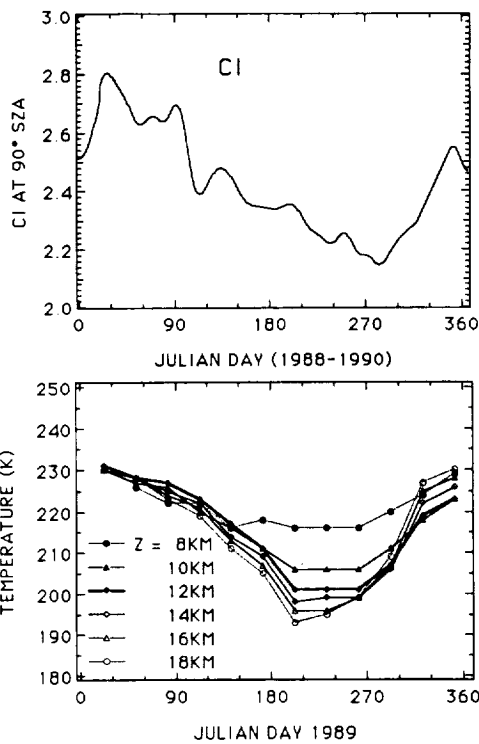


Figure 4. Three years average seasonal variation of the sky color index at  $90^\circ$  SZA and monthly mean temperature in the lower stratosphere above Dumont d'Urville. The bluer twilight sky at Spring than in Fall, could be partly caused by the formation of cirrus at or above the tropopause.

#### After volcanic eruptions

The picture is slightly different in 1991 (figure 1), where larger blueings occurred since around day 215 (early August), followed later by unusual reddenings highly correlated to the 50 hPa temperature variations, indicative of the motion of the polar vortex (figure 5). Figure 6 similar to figure 2, but for the end of 1991, shows that both blueings and reddenings can occur at warm temperature. Therefore they are not PSC. According to the model, blueings would result from a dense absorbing layer in the lower stratosphere, and reddenings at higher altitude, the last being observed outside the vortex only.

Lidar measurements performed at Dumont d'Urville during favourable weather periods (Godin et al., same proceedings) show that two layers were present above the station: one almost permanently around 10-14 km (optical thickness 0.06) and the second more sporadic and variable between 22 and 26 km (optical thickness 0.03). The first is thought to result from the eruption of Mt Hudson in Chile in August, and the second from the one of Mt Pinatubo in Philippine in June.

Figure 7 shows the result of inclusion of these layers into the model. The low altitude layer makes the twilight color index to shift toward the blue while the high altitude layer would shift the color index toward the red, in agreement with the experimental data.

SAOZ daily observations give the details of the evolution of the two layers. The low altitude Hudson cloud arrived above Dumont d'Urville in early August, that is rapidly after the eruption and penetrated into the vortex, which is then not closed at altitudes around 10-14 km. The high altitude Pinatubo layer was observed above the station at the time of the first warming only (day 248-249, 4-5 September) and did not penetrate into the polar vortex, which appears then to be isolated at an altitude of 22-26 km.

## 5. CONCLUSION

Zenith sky UV-visible spectrometry at twilight, able to derive the daily variation of stratospheric scatterers from a color index, and lidar, able to measure their vertical distributions but during clear days only, appear to be a powerful combination for monitoring stratospheric aerosol and PSC.

The four years of spectrometric data available now at Dumont d'Urville shows that dense PSCs are rather unfrequent above this station located at the edge of the vortex. They demonstrate also the formation at the end of the winter above Antarctica, of a scattering layer at 8-12 km in the lower stratosphere, which causes the color of the twilight sky to be bluer at spring than in Autumn.

They demonstrate also that volcanic aerosol which arrived above the station in August-September 1991, penetrated rapidly into the polar vortex at 10-14 km, but not at 22-26 km.

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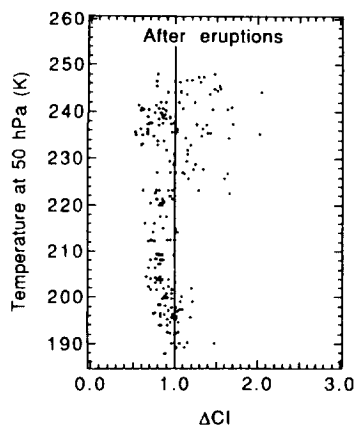


Figure 6. Relation between  $\Delta CI$  and 50 hPa temperature after August 1991. Large temperature independent departures compared to the previous years average are observed. Persistent blueings are caused by low altitude Mt Hudson aerosol layer. Large reddenings are caused by the high altitude Mt Pinatubo aerosol layer.

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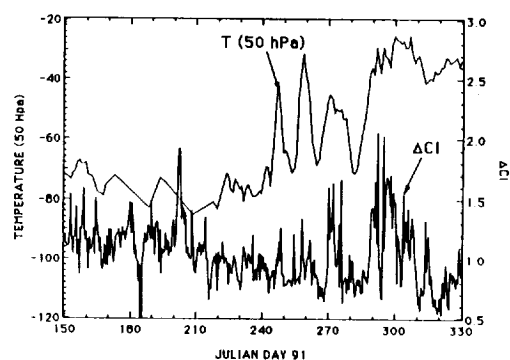


Figure 5. Color index variation at twilight ( $\Delta CI$ ) and 50 hPa temperature observed from June until November 1991. An average shift toward the blue is observed after day 215 while large reddenings occurred later, highly correlated with stratospheric warmings.

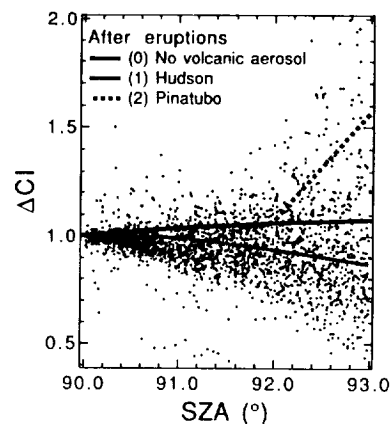


Figure 7. Color index variation observed at twilight at the end of 1991 and model calculations with aerosol distribution observed by lidar.