Observations of absorbing layers in the Antarctic stratosphere in October 1991

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SUMMARY

Measurements of the transmission of solar infrared radiation through the earth’s atmosphere, by the HALOE experiment on the UARS spacecraft, were made at high southern latitudes during October 1991. These observations are direct measurements of atmospheric transmission, and do not need to be passed through a composition or temperature retrieval process; they are, therefore, amenable to direct interpretation. During October 1991 the profiles of transmittance versus height in the atmosphere show clear evidence for the arrival (from more northerly latitudes) of layers of some material which absorbs infrared radiation, at heights of up to 28 km. The spatial structure of the absorbing material shows considerable variability with longitude at a given latitude. The spectral properties of the detected absorption, measured at the various HALOE wavelengths, are consistent with absorption by sulphate aerosol, and clearly implicate the volcanic eruptions from Mt Pinatubo and Hudson during 1991. These results provide direct and detailed evidence for the arrival of layers of what appears to be sulphate aerosol during the 1991 southern spring, at latitudes as high as 80 degrees south.

1. INTRODUCTION

HALOE is a limb-sounding infrared solar occultation radiometer on board the Upper Atmosphere Research Satellite, UARS, which was launched on the Space Shuttle Discovery on 12 September 1991. The experiment employs techniques of gas correlation radiometry to measure the concentrations of atmospheric hydrochloric acid (HCl), hydrofluoric acid (HF), nitric oxide (NO) and methane (CH₄), and broad-band filter radiometry to measure nitrogen dioxide (NO₂), water vapour (H₂O), ozone (O₃) and temperature—through measurements in the carbon dioxide (CO₂) 2.8 μm band—in the mesosphere, stratosphere, and upper troposphere. Note that the gas correlation channels also provide a broad-band filter measurement; these broad-band observations are used in the analysis which follows. A full description of the HALOE experiment may be found in the paper by Russell et al. (1993a). Early observations of the southern spring vortex have been reported by Russell et al. (1993b).

UARS is in a 57° inclination, drifting-phase orbit, at an altitude of 585 km. There are 15 orbits per day, and therefore 30 occultations of the sun by the limb of the atmosphere, which are grouped into 15 sunrise events distributed on a circle of near-constant latitude, and 15 sunset events similarly grouped on another latitude band. The latitudes of the sunrise and sunset events vary with time, as illustrated in Fig. 1; occasionally sunrise and sunset events coincide in latitude. The maximum latitudes of observation reached, defined by a combination of orbital inclination and limb-view direction, are 80°N and S. HALOE began atmospheric observations on 11 October 1991, and measurements at high southern latitudes continued through October and November of that year.

The raw measurements made by HALOE comprise detected solar signal voltages as a function of limb altitude (or pressure level) as the sun rises or sets behind the atmospheric limb. These voltages are normalized to give an atmospheric transmittance

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by dividing them by the measured exo-atmospheric solar signal voltage. It is the transmittances, $\tau$, that are the subject of this paper. Because the measurements are direct, and do not pass through a further retrieval process as is the case in deriving atmospheric temperatures or constituent concentrations, they may be directly interpreted, with relatively few sources of error being introduced in the processing. This is of some advantage because, however precise a retrieval process may be, it is bound to introduce additional errors (e.g. due to uncertainties in spectral-line parameters).

In this study, direct measurements of the transmittance of the atmosphere, averaged over the spectral pass-bands of the HALOE experiment channels, are reported. These measurements show clear evidence of discrete absorbing layers of some material moving into a previously relatively transparent Antarctic stratosphere during October 1991. Both spatial and spectral analyses of the data throw light on the composition of these absorbing layers, as well as on aspects of the dynamical control of the Antarctic circumpolar atmosphere in the austral spring.

There have been numerous reported observations of both aerosols and polar stratospheric clouds (PSC) in the stratosphere. As an example, a recent publication (Rosen et al. 1992) describes observations made with a balloon-borne two-wavelength backscatter instrument between October 1991 and March 1992, which show the arrival of Pinatubo aerosol layers over the Arctic, at Alert (82.5°N, 62.5°W) and Resolute (74.7°N, 95.0°W), during the northern winter/spring. Layers of aerosol at heights up to 20–22 km were observed over Alert in December 1991 and January 1992. Deshler et al. (1993) report measurements of Pinatubo aerosol vertical profiles and size distributions at 41°N during 1991 and 1992; and Winker and Osborn (1992) made LIDAR measurements of Pinatubo aerosol in the equatorial zone in July 1991. Collins et al. (1993) have reported observations of LIDAR scattering from polar stratospheric clouds in the southern hemisphere, at altitudes of between 12 and 27 km, as the stratosphere cooled during the winter from late May 1990 to late August of the same year. Measurements by Adriani et al. (1992) in late August 1991 using LIDAR and balloon-borne particle counters indicated both

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**Figure 1.** Example of variation of the latitude of observations with time for both sunrise (circles) and sunset (crosses) events: 1 year, beginning 4 October 1991.
water ice and nitric acid trihydrate ice clouds between about 10 and 23 km; these authors noted that no PSCs were observed after the end of August 1991. Measurements of a more global nature from the SAGE II experiment are reported by Trepte et al. (1993), who observed the southward extension of the Pinatubo aerosol cloud (determined by 1 μm extinction ratios) passing 60°S (the highest southern latitude observed) between early October and early November. We shall see below that this is consistent with the present study. Thomason and Poole (1993) discuss the use of SAGE II extinction measurements of aerosols as a diagnostic of vortex processes. Charlson and Wigley (1994) discuss the overall importance of sulphate aerosol to the climate.

2. THE OBSERVATIONS—DETECTION OF ABSORBING LAYERS

All the data in this paper will be expressed in the form of $\tau(\lambda, z)$, that is transmittance as a function of wavelength $\lambda$ (=HALOE channels), and of altitude, $z$, at the limb of the atmosphere (strictly, the tangent-point altitude—see Russell et al. 1993a). Data are

![Figure 2. End-to-end spectral bandpass of all the HALOE channels.](image-url)
reported for all channels, though in the cases of HF, HCl, NO and CH₄ only from the wide-band signals, since these are more sensitive to broad-band absorptions than are the gas filter channels.

Figure 2 and Table 1 describe the spectral properties of the wide-band HALOE channels. It must be remembered that the observed transmittances are averages over wavelength, weighted by these filter profiles. In general, for molecular absorptions, the curve of \( \tau (\lambda, z) \) versus altitude, \( z \), should be a smoothly varying exponential-like curve, tending to unity at the top of the atmosphere, and to zero in the troposphere. Relatively narrow structure with height on such curves is not expected to arise from gaseous absorbers, except where, for example in the case of \( O_3 \), the density distribution may be strongly layered.

High-latitude data were obtained on only a limited number of days in October 1991, for operational reasons. Data used for this study were obtained on 11, 12, 18, 23, 24, 26, 27 and 29 October. On each of these days observations were made at various longitudes around the earth, in the latitude bands indicated in Table 2.

<table>
<thead>
<tr>
<th>Species</th>
<th>Gas filter/wide band</th>
<th>Spectral range, cm⁻¹ (microns)</th>
</tr>
</thead>
<tbody>
<tr>
<td>HF</td>
<td>GF + WB</td>
<td>4019–4136 (2.45)</td>
</tr>
<tr>
<td>CO₂</td>
<td>WB</td>
<td>3508–3636 (2.80)</td>
</tr>
<tr>
<td>HCl</td>
<td>GF + WB</td>
<td>2886–2994 (3.40)</td>
</tr>
<tr>
<td>CH₄</td>
<td>GF + WB</td>
<td>2850–2932 (3.46)</td>
</tr>
<tr>
<td>NO</td>
<td>GF + WB</td>
<td>1869–1931 (5.26)</td>
</tr>
<tr>
<td>NO₂</td>
<td>WB</td>
<td>1586–1614 (6.25)</td>
</tr>
<tr>
<td>H₂O</td>
<td>WB</td>
<td>1500–1528 (6.60)</td>
</tr>
<tr>
<td>O₃</td>
<td>WB</td>
<td>957–1080 (10.04)</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Date October</th>
<th>Latitude (°S)</th>
</tr>
</thead>
<tbody>
<tr>
<td>11</td>
<td>75.5–75.7</td>
</tr>
<tr>
<td>12</td>
<td>75.7–75.8</td>
</tr>
<tr>
<td>18</td>
<td>79.8–79.9</td>
</tr>
<tr>
<td>23</td>
<td>76.1–75.8</td>
</tr>
<tr>
<td>24</td>
<td>74.9–74.7</td>
</tr>
<tr>
<td>26</td>
<td>72.1–71.7</td>
</tr>
<tr>
<td>27</td>
<td>70.7–70.4</td>
</tr>
<tr>
<td>29</td>
<td>67.2–65.8</td>
</tr>
</tbody>
</table>

Figure 3 illustrates two \( \tau (\lambda, z) \) curves, selected to demonstrate the clear observation of a discrete absorbing layer at one longitude but not at another at essentially the same latitude. In this case the value of \( \lambda \) is 3.40 μm (HCl channel—see Table 1) and the two observations were made on 12 October 1991, at positions 75.7°S, 37.8°E and 75.8°S, 325.1°E. In the former case there is quite clear evidence of an intense layer of absorption at about 26 km altitude, but no such evidence exists in the latter case, which looks much more typical of the exponential curve that would be expected for absorption by a gaseous absorber such as CH₄, which is the primary gas affecting the broad-band signal in this channel.
If we compare these two cases with the observations made at the same places and times, but in a different HALOE spectral channel (Fig. 4: in this case the water vapour channel at 6.6 µm) we see that very little evidence for an absorbing layer is detected. Examination of other channels shows that there is a strongly varying spectral signature associated with this absorption. Thus, we can conclude initially, in qualitative terms, that there is evidence of absorbing layers at high altitudes and high southern latitudes, which shows an apparent dependence both on geographic location and on spectral wavelength. We shall now go on to examine these dependencies in detail.

3. DEPENDENCE OF TRANSMITTANCE ON THE SPECTRAL CHANNEL

Figures 5(a) to (d) show a series of observations, made on 11 October 1991, at two positions, 75.7°S, 86.2°E and 75.7°S, 62.0°E, for all the HALOE channels in order of
Figure 5. Transmittance of the atmosphere during sunrise on 11 October 1991 at two positions, 75.7°S, 86.2°E (solid curves) and 75.7°S, 62.0°E (dotted curves). (a) Top: at a wavelength of 2.45 μm (HF); bottom: 2.80 μm (CO₂). (b) Top: 3.40 μm (HCl); bottom: 3.46 μm (CH₃). (c) Top: 5.26 μm (NO); bottom: 6.23 μm (NO₂). (d) Top: 6.60 μm (H₂O); bottom: 10.04 μm (O₃).
Figure 5. Continued.
increasing wavelength. It was found that any variability of absorption strength observed as a function of location is independent of the variability as a function of wavelength. From an examination of the data, it can be seen that at 2.45 μm (HF channel) a detectable layered absorption is observed (Fig. 5(a)) at an altitude of between 25 and 28 km; however, no significant layer absorption is detected at these heights at 2.80 μm (CO₂ channel)—although, note that the strong background due to molecular absorption masks a possible layer at lower altitude of about 23 km. The strongest layer absorptions occur in the channels at 3.40 (HCl channel) and 3.46 μm (CH₄ channel) (see Fig. 5(b)), and the absorption has again become weaker by 5.26 μm (Fig. 5(c), top frame—NO channel). At 6.25 μm (NO₂) and 6.60 μm (H₂O) (Fig. 5(c), lower frame; Fig. 5(d), upper frame), no significant layer absorption is observable. At 10.04 μm, the O₃ channel (Fig. 5(d), lower frame), it would appear that there is again a significant absorption difference between the two curves shown, although in this case it is suspected that the known high variability in ozone around a circle of latitude is contributing to the observed differences.

These observations are consistent with a more complete analysis of the observed dependence of aerosol absorption on wavelength in the HALOE data, which has been

![Diagram](image_url)

Figure 6. Wide-band transmittance in the HALOE CH₄ channel observed near 76°S on 11 October 1991: (a) at longitudes between 231 and 304°E; (b) at longitudes between 62 and 159°E.
reported by Hervig et al. (1993), the results of which can be briefly summarized here. Hervig et al. have described how such spectral dependence has been used quantitatively to compare the observed aerosol absorption with theory. To do this, the transmittance at the level of maximum absorption in a layer, \( \tau_p \), has been derived from the ratio of highest and lowest measured transmittances, measured around a circle of latitude (for example by the ratio of transmittances at 26.2 km in Fig. 3). The extinction coefficient, \( \beta \) (in units of \( \text{km}^{-1} \)), is then defined by the relationship \( \tau_p = \exp (- \int \beta ds) \), where \( s \) represents path length (along the limb path) in the atmosphere. The term \( \beta \) is actually derived by Hervig et al. by an ‘onion-peeling’ technique, where the path length is modelled from the known sun-satellite geometry. This result is then compared with a model which describes the absorption due to a sulphate aerosol, assuming spherical particles, and a single mode log-normal size distribution. The size distribution is taken from balloon measurements made over Wyoming in August 1991 (Deshler et al. 1993). It is shown by Hervig et al. that the maximum in absorption observed by HALOE at about 3.5 \( \mu \)m is consistent with the layers being composed of sulphate aerosols.

The time of arrival of these layers over the Antarctic in October 1991 reported here suggests that the aerosols may be the product of the eruptions of Mt Pinatubo in the Philippines, at position 15.1°N, 120.3°E (15 June 1991), or of Mt Hudson in the Chilean Andes, at position 45.9°S, 287.0°E (15 August 1991). The fact that meridional transport in the stratosphere is fast enough to move these aerosols to the Antarctic by October is confirmed by the measurements of stratospheric optical depth by the SAGE II instrument, made at latitudes between about 70°N and S, reported by Trepte et al. (1993).

The question arises as to whether the observed absorptions could be due not to aerosols, but to polar stratospheric clouds (Toon et al. 1989; Hanson and Mauersberger 1988). This possibility seems to be ruled out by the temperatures prevailing at about 25 km on the days in question, which were in the range 210–220 K, considerably higher than those characteristic of PSC formation (WMO 1989). Also, laboratory work (e.g. Tolbert and Middlebrook 1990), indicates that for the pure water ice and highly hydrated nitric acid (e.g. nitric acid trihydrate) thought to make up type 1 and type 2 PSCs, there may be a peak in absorption, but at a shorter wavelength (c. 3.1 \( \mu \)m) than observed in the present data. For both these reasons, it seems reasonable to rule out PSCs as the cause of the observed layer absorption.

4. Dependence of Absorption on Location and Time

The examples of HALOE data shown earlier in this paper have shown how the presence of the absorbing layers seen over the Antarctic in October 1991 depend strongly on location. At a given latitude, typical of a single day of HALOE observations, absorption by the layers was present at some longitudes but not at others. In this section we examine the implications of this spatial variability.

Figures 6 and 7 show the transmittance of the atmosphere between altitudes of 15 and 30 km in the HALOE methane channel (3.46 \( \mu \)m) during a number of occultation events on both 11 and 26 October 1991. Figure 6(a) refers to 11 October, a mean latitude of about 75.5°S, and longitudes between 231 and 304°E. Figure 6(b) refers to a different arc of latitudes, namely 62 to 159°E, on the same day. Figure 7(a) shows data for 26 October, a mean latitude of 72.2°S, and a longitude range of 205 to 302°E; while Fig. 7(b) covers the longitude range 60 to 181°E for the same day. These figures illustrate the trends that were observed during October: firstly, the atmosphere at high latitudes showed an increase in the frequency and intensity of absorption events in the longitude sector included in the range 60 to 180°E as time evolved through the month; secondly,
the sector between 230 and 305°E remained quite clear of absorption during this period; and, thirdly, the altitude of the peak absorption fell from about 25 km on 11 October to nearer 20 km by 26 October (however, it is apparent from Figs. 6 and 7 that the transmittances measured at levels above 25 km did not in fact change significantly, but rather the absorption at lower levels increased, indicating perhaps not a fall in the altitude of the topside of the layers but the later arrival of absorbing material at lower altitudes). On later dates, into November and later, when the HALOE observations moved to lower latitudes, very high absorption became the norm in the lower stratosphere at all latitudes, longitudes and altitudes sampled by HALOE. This trend was presumably due to the gradual dispersion of volcanic aerosol to higher latitudes with time, away from the lower latitude source region, helped by the subsequent seasonal breakdown of the polar vortex, and associated mixing of air from lower latitudes.

The understanding of how lower latitude air mixes with so-called ‘polar-vortex’ air is a complex and still controversial subject, with theories ranging from a highly self-contained vortex ‘containment vessel’ to the possibility of large-scale transfers of air through a permeable ‘chemical processor’ over the southern pole. It is not the purpose

Figure 7. Wide-band transmittance in the HALOE CH₄ channel observed near latitude 72°S on 26 October 1991: (a) at longitudes between 205 and 302°E; (b) at longitudes between 60 and 181°E.
in this paper to argue in support of one model over another; however, we can attempt
a limited interpretation of the results presented here with the aid of analyses of the
dynamical situation over Antarctica during October 1991. The reader will need to refer
to the latitudes of observations listed in Table 2.

During the second half of the month, the vortex was considerably distorted and was
centred approximately at the edge of the Weddell Sea, at about position 75°S, 310°E.
The meteorological situation on 22 October is illustrated in Fig. 8, which shows the 30 mb
(about 24 km) wind fields at 12.00 GMT, together with a series of crosses which mark the
position of the HALOE observations on that day. The date 22 October was chosen as
being roughly midway between the 11th and the 26th, and the situation did not change
radically during the whole period. The figure makes it clear that the zone from roughly
0 to 240°E coincides with a region where wind speeds are high (30–40 m s⁻¹). The zone
from about 260 to 360°E corresponds to much lower wind speeds, nearer the centre of
the vortex. An investigation of Figs. 6 and 7 shows that, indeed, while the frequency of
absorption events, and the amount of absorption both increased between the 11th and
the 26th of the month, this happened almost exclusively in the latitude sector covered
by Figs. 6(b) and 7(b), i.e., between about 60°N and 180°E. Comparison with Fig. 8 shows
that this is a region where the HALOE observations were well away from the vortex
centre. Without wishing to overinterpret these data in terms of penetration or not of
vortex boundaries and jet streams, we can say non-controversially that we would expect
this sector of high winds to experience the arrival of any extra-polar material before air

![Figure 8](image_url)

Figure 8. The analysis of the meteorological situation on 22 October 1991, at 30 mb (24 km): the arrows
represent local wind vectors. The Greenwich meridian runs from the centre to the bottom of the diagram, and
the latitude circles are drawn at 50 and 75°S. Longitude lines are spaced by 20°. Strong absorption occurs in
the longitude sector from about 60 to 180°E, while weak/no absorption occurs in the sector 230 to 310°E. The
large crosses indicate the positions of HALOE observations.
in the vortex centre. Conversely, the profiles for which little or no evidence exists for layer absorption all lie in the longitude range 230–310°E. From Fig. 8 we can see that this coincides with air that is very closely associated with the centre of the vortex, air which presumably has experienced little exchange with lower latitudes for some time.

The coincidental facts that the HALOE experiment samples on circles of latitude symmetrically disposed around the pole (Table 2), while in October 1991 the polar vortex was considerably displaced from the pole, at a time when the relatively clean air of the Antarctic stratosphere was being ‘contaminated’ by volcanic aerosol, seems to have provided us with a beautiful natural experiment. In the circumstances, we were able to use HALOE to sample clean air near the centre of the vortex, and air near the edge; the air at the edge showed the encroachment of aerosol contamination. Later in the year, after the breakdown of a recognizable vortex, with air well mixed at all latitudes, the transmittance in all HALOE channels became very low at altitudes below about 25 km at all locations sampled by HALOE (i.e. up to 80°S). It must be concluded that by this time the Antarctic lower stratosphere was fully ‘contaminated’ by aerosol.

5. CONCLUSIONS

Measurements of the vertical profile of atmospheric transmittance at a number of infrared wavelengths, made by the HALOE experiment on the UARS spacecraft, have been analysed to show that during October 1991 there was evidence for the detection of discrete absorbing layers at high southern latitudes in the lower stratosphere. The absorption by these layers has been shown to be strongly wavelength dependent, and observations between 2.45 and 10.05 μm by HALOE have been used to investigate the composition of the absorber. A peak in absorption occurs at about 3.5 μm and this is consistent with other studies which indicate that the absorber is sulphate aerosol. This is suggestive that the origin of the absorbing layers is in the volcanic aerosol produced by Mts Pinatubo and/or Hudson earlier in 1991. It is known that effluent from Pinatubo reached heights above 25 km, while that from Hudson reached up to about 18 km.

HALOE provides 15 sunrises and 15 sunsets per day, arranged around nearly constant circles of latitude. In mid October 1991, sunrise events were sampled to about 75°S, while in later parts of the month, sunset events were obtained to about 79°S. Daily observations taken around circles of latitude indicated that the absorbing layers were present at some longitudes but not at others. An inspection of the meteorological situation in the second half of October indicates that the polar ‘vortex’ was significantly displaced towards South America (approximately 15° off the pole) and that the circle of HALOE observations was, in fact, sampling both the centre and other regions inside and near the edge of the vortex. Air at the centre remained ‘clean’ throughout October (i.e. showed no sign of the absorbing layers). Air nearer the edge of the vortex showed high absorption episodes due to the contaminant layers of absorber. Thus it would appear that the sampling of the HALOE polar-symmetric experiment and the polar-asymmetric displacement of the vortex, at a time when volcanic aerosol was spreading poleward, provided the circumstances under which sampling both near the vortex centre and at the edges was possible.

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