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SIMULTANEOUS STRATOSPHERIC AEROSOL AND OZONE LIDAR MEASUREMENTS AFTER THE PINATUBO VOLCANIC ERUPTION

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Abstract. Preliminary results of simultaneous ozone and aerosol lidar measurements taken in the period July-early October 1991 are presented. The main problem for retrieving the ozone profile is the correction of the *on* and *off* DIAL signals. The backscattering ratio obtained by the *off* signal is used for this purpose and ozone profiles are validated against ozonesonde data. A number of cases are presented including a few occasions of layers with large backscattering ratio. For these events the differences between ECC sonde and DIAL range between 5% to 20% and indicate that the method adopted for correction needs improvements especially for very thin layers. Sensitivity tests show that the retrieved ozone may change by roughly 10% depending on the choice of the aerosol optical parameters. Absolute difference between ECC and DIAL could not be completely explained in terms of the uncertainty in those parameters. The data obtained may still be used to prove the effects of heterogeneous chemistry on ozone destruction rate. Given the uncertainties introduced by the aerosol correction large amount of data need to be accumulated.

Introduction

Large volcanic eruptions may produce considerable amount of sulfate aerosols in the stratosphere. Until a few years ago the main interest for these events was their possible influence on climate (Robock, 1991) and the use of the aerosols as a tracer for the stratospheric circulation. After the suggestion make by Hofmann and Solomon (1989) volcanic aerosols may also be responsible of a considerable ozone depletion, based on the fact that aerosol surface may accelerate the conversion of nitrogen pentoxide to nitric acid. The shift of the NO_x, NO_y balance would result in a slower rate of formation for chlorine nitrate and would make available more free chlorine to destroy ozone. Hofmann and Solomon have shown that evidence to substantiate such a mechanism already existed for the El Chichòn eruption.

The eruption of volcano Pinatubo around the middle of June of 1991 may provide a critical test for this theory and it is of paramount importance to have data on both the evolution of the stratospheric aerosol load and ozone. Aerosol lidar have provided data on the optical thickness after volcanic eruptions that can be simply related to the area density of aerosols. This data is the most important (along the sticking coefficient) to evaluate the aerosol effects on ozone. Lidar in its implementation as DIAL (Differential Absorption Lidar) may also provide ozone profiles in the stratosphere. However in presence of large amount of aerosols, the DIAL signals are disturbed and require a suitable correction. Several ways have been suggested to this purpose, as in Browell et al. (1985): using a DIAL system they show the sensitivity of the retrieved tropospheric ozone to aerosol parameters for backscattering ratios of the order of 1 to 3. Uchino and Tabata (1991) give a simple algorithm to correct stratospheric ozone in presence of very small scattering ratios. The main problem however is to determine a correct backscattering ratio that can be accomplished following the suggestion of Klett (1985) and implemented by Browell et al. (1985). The method, which we use, however also includes ozone absorption.

In this paper we report simultaneous measurements taken with an aerosol lidar and an ozone DIAL following the eruption of the Pinatubo volcano. The ozone data are validated in several occasion with ECC soundings to estimate the reliability of the correction method. This correction requires assumptions about optical parameters of the aerosols. We perform a number of sensitivity tests on the choice of these parameters to assess their effect on the retrieved ozone.

Lidar Observations

The lidar station of the University of L'Aquila (42N, 13E, 700m a.s.l.) includes an aerosol lidar system as described in D'Altorio and Visconti (1983) with a more powerful pulsed dye laser (> 1 J *per pulse*) operating at about 0.15Hz. Recently we started to operate a DIAL system using an excimer laser emitting at 308nm and 351nm with typical output energies of 90mJ and 70mJ respectively and a repetition rate ranging between 1Hz and 80Hz. The two laser beams are mixed with a system of dichroic mirrors and the backscattered signal is collected with a f/10, 1m Cassegrain telescope. A detailed description of the system is given in Masci et al., (1991). In a typical measurement session (3 to 4 *hours*), 800 to 1000 laser shots are accumulated in the visible channel and 5×10^4 to 25×10^4 *shots* on the DIAL system. It has to be considered that during the period to which these measurements are referred we had unusually bad weather conditions and also clear nights were characterized by some haze which resulted in the degradation

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of the received signals. For the aerosol lidar the signal was averaged over $0.3km$ while for the DIAL a running mean was performed over $3km$ range with $0.3km$ step. The ozone profile was recovered with an algorithm similar to those described in Uchino and Tabata (1991) and with more detail by Godin (1987). At the present time we are still working on a precise estimate of the errors affecting the retrieved ozone profile, but we expect them to be in the range of a few percents at the lowest altitudes, raising to about $8-10\%$ at the upper boundary.

Validation of the DIAL results can be made against independent measurements, typically ozonesonde. In Italy the only site operating an ozonesonde is near Bologna $\sim$ $300km$ north of our lidar site. We have made such intercomparison whenever possible even if the measurements were taken within few days of the sounding. The first evidence at our site of the aerosol presence was obtained on August 7 as shown in Fig.1. However the system could not be operated during the second half of July due to weather and technical problems, but there is possible evidence from the CNR lidar in Frascati near Rome of a small scattering ratio (~ 1.2) near $15km$ from July 17 (Congeduti F., private communication 1991). The first intercomparison with ECC data was made with a DIAL profile taken on July 1st probably unaffected by Pinatubo aerosols (Masci et al., 1991). Starting at the end of July up to the time this paper is written, data were taken whenever possible, and the results shown in Fig.2 refer only to those days in which ECC sounding were also available. Data on the visible channel are fitted in the upper part of the range with a simulated molecular atmosphere which however takes into account the ozone absorption due to the Chappuis band and the aerosol extinction with a self-consistent algorithm. As shown in Fig.1 the backscattering ratios stay quite constant in the $15km$ range, while occasionally some layers appears above $20km$. Since the end of September a large aerosol layer was observed in the 18 to $25km$ range. The backscattering ratios at the DIAL channels were also obtained in the same manner of the visible one. The layers features are quite well reproduced in all three wavelengths. The DIAL wavelengths show roughly the same backscattering ratios ($R(\lambda, z)$ with λ wavelength and z altitude) when these are small (i.e. 1.2).

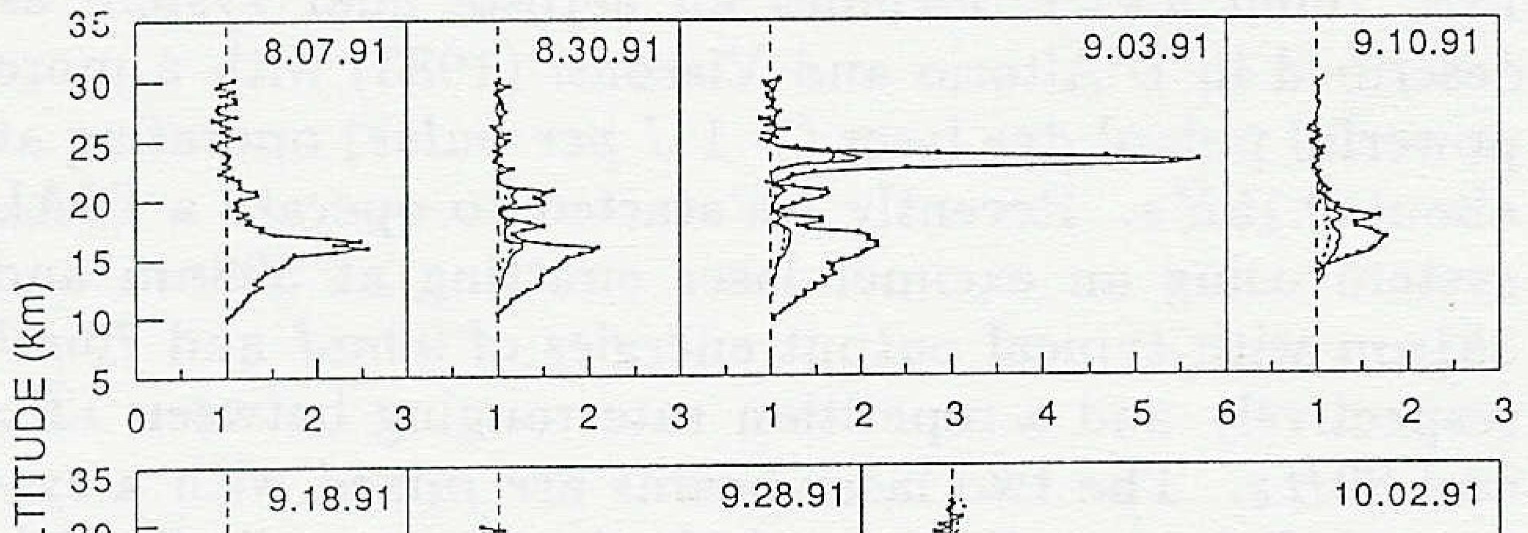

Fig.1. Lidar backscattering ratios from August to October 1991 for three wavelengths. Solid lines with dots refer to $590nm$, solid lines to $351nm$ and dashed line to $308nm$.

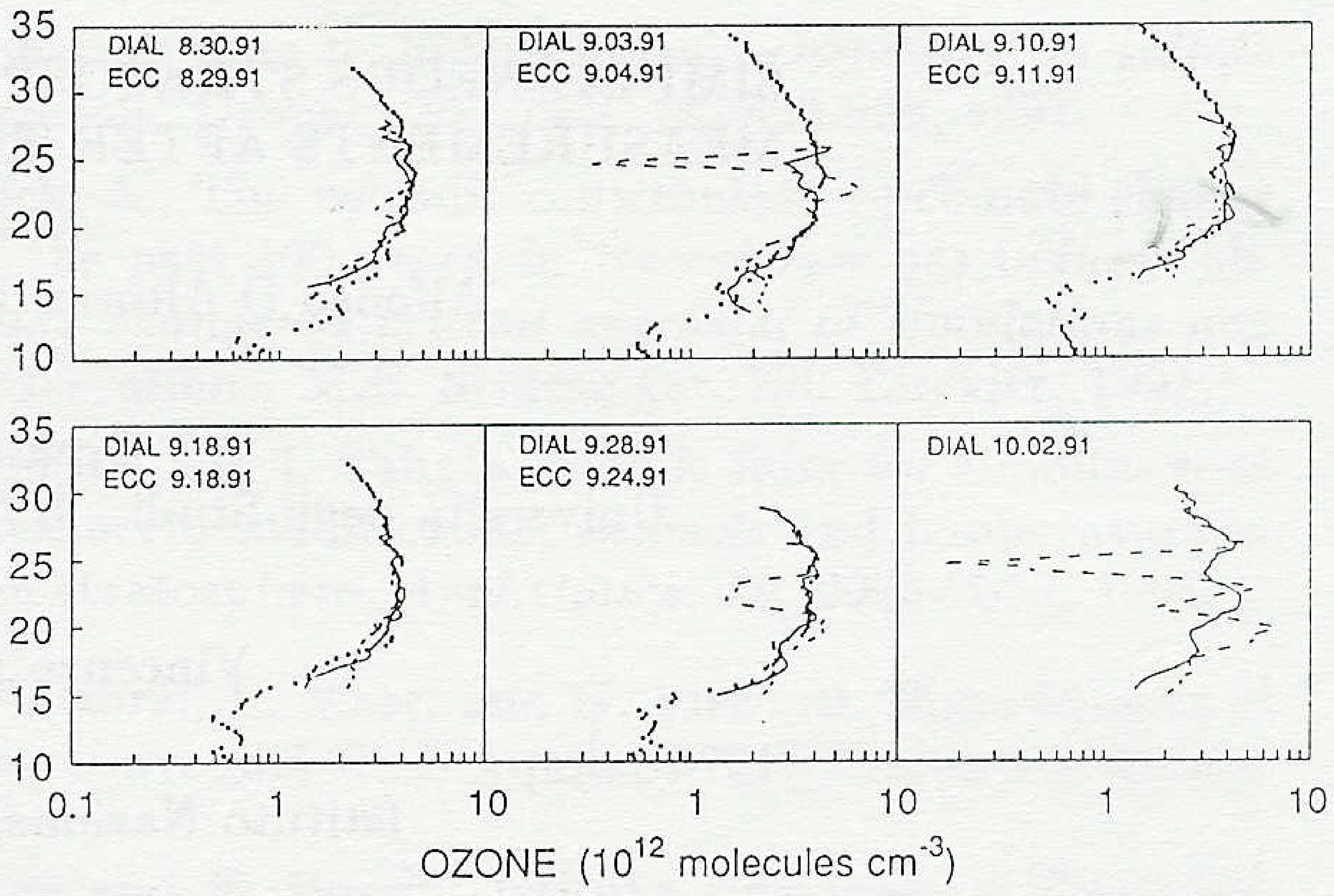

Fig.2. Ozone profiles for the same days of Fig.1 except August 7. Dashed lines are the uncorrected profiles from aerosols while the correct ones are shown with the solid lines. The data from the ECC sonde (dots) are also shown except for October 2 when data are not available.

This may be due to the uncertainties associated with the fitting process or to the different size distribution for the lowest aerosols. On September 3 our lidar detected a very strong layer around $23km$ with backscattering ratio around 5.5, a similar situation we had after September 28. As shown in Fig.1 $R(\lambda, z)$ scales down correctly along the three wavelengths, for the upper layer, but again not for the lower layers which have smaller backscattering ratios. The DIAL ozone tends to give smaller values above $25km$ when compared to ECC data. We attributed this effect partly to our signal becoming noisier above that altitude but also to systematic errors introduced by ECC sounding (Kirchoff et al., 1991).

The most delicate part of these preliminary results is the correction of the ozone to such a degree and precision to extract effects related to the presence of sulfate aerosols. The procedure is to get the backscattering ratio for a wavelength in which the ozone absorption is negligible and to scale it to the other wavelengths. The visible channel in our case has the advantage to maximize the scattering from aerosol with respect to molecular one but has an ozone absorption larger than the *off* DIAL wavelength. In addition this is closer to the *on* wavelength and samples the same air parcel at the same time. We have used both wavelengths to correct the *on* signal and will present here only the results obtained with the $351nm$ which seem more satisfactory. An accurate analysis of this problem is beyond the purpose of this paper and needs in any case a much larger amount of data than the one in our hand at the present time. As shown by Browell et al. (1985), the wavelength dependence of the backscattering cross section may be taken as λ^{-m} where the value of m varies usually from 0 to 2. Then, the backscattering mixing ratio, $R(\lambda, z)-1$, can be scaled as λ^{4-m}. The lidar return at $308nm$ is corrected starting from the backscattering ratio at $351nm$. All the ozone data were reduced with this procedure and a sample is presented in Fig.2. It appears that the correction works quite well and help to reconcile the DIAL data with the ECC in those regions where $R(\lambda, z)$ is large. This is particularly evident in the case of intense layers like those of September 3 and 28 and October 2. A more accurate comparison can be made in terms of the difference between the DIAL ozone and the ECC data as shown in Fig.3 for three representative profiles. The correction is very good for moderate scattering, but may still present problems for very intense and isolated layers as those of September 3 and 28. It is to notice however that these comparisons refer to ECC data taken within 24 hours and not in the same site so that we could always expect differences due to natural variability. As stated above these corrections may be sensitive to the choice of two parameters: the extinction to backscattering ratio, C, and the value of m (defined above). A sensitivity test has been carried out by changing m from 0.8 to 1.2 and C from 20 to 40 for the September 3 event and the results are shown in Fig.4. The largest changes on the retrieved ozone ($\sim$ 5%) are observed just below and above the aerosol layer and are symmetric with respect to values $m = 1$ and $C = 30$. However changing C will affect mainly the lower part of the ozone profile because there the optical thickness is larger. The fact that the uncertainties in these two parameters could affect the ozone profile only up to 10% is quite important when considering that the expected local ozone depletion may be of the order of 10 - 15% as results from preliminary calculation, which have been made with a 2D model for a similar event (Pitari et al., 1991). As shown in Fig.3 however the residual between ECC and DIAL may exceed 20% of the observed value and that could not be attributed to the choice of m or C. Other cases reported in the same figure do show a much smaller residual. Notice that Browell et al. (1985) reach the same conclusions.

As discussed above an evaluation of the m factor could be obtained with some accuracy especially in the case of large backscattering ratio as those of September 3 and 28 and October 2. From this preliminary analysis values for m ranging from 0.8 to 1.3 were obtained.

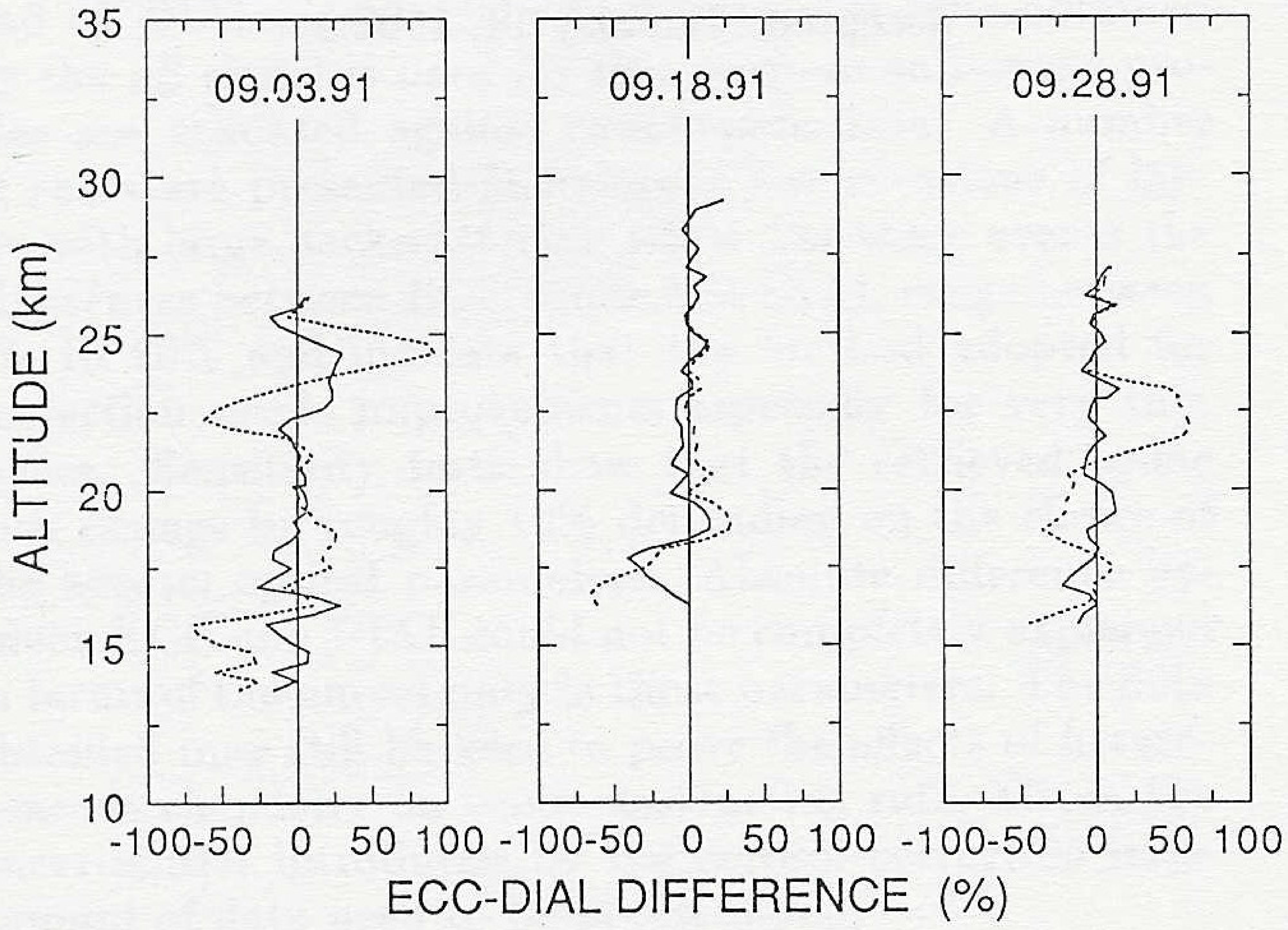

Fig.3. Difference between the DIAL and ECC ozone (percentage in respect of ECC profile) for the days shown. Differences with the uncorrected and corrected ozone profile are shown with dashed and solid line respectively. Notice that September 3 and 28 presented large backscattering ratio.

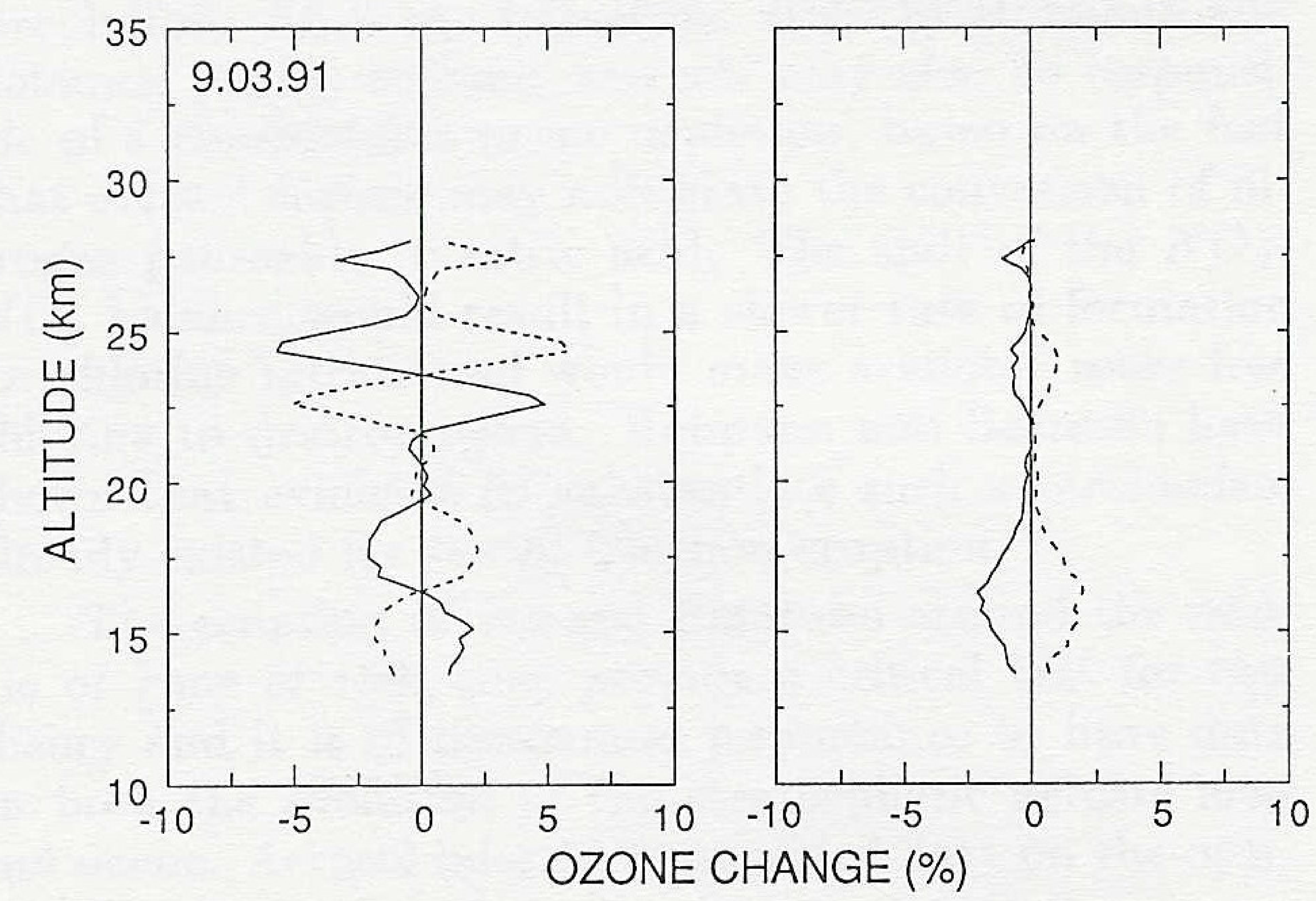

Fig.4. Sensitivity of the corrected ozone values to the assumption of parameter m (see text) and the extinction to backscattering ratio, C, for the aerosols. For m the percentage change are referred to $m = 1$ while for C to C=30.

Conclusions

Preliminary results obtained for the DIAL ozone profiles in presence of strong aerosol layers show that corrections are possible even considering the uncertainties in the aerosol optical parameters. This conclusion is supported by a comparison between ECC sounding values and the DIAL ozone profile; also sensitivity tests, carried out with a large excursion of the extinction to backscattering ratio, C, and of the exponent m, show that the indetermination introduced in the retrieved ozone profile is comparable to the expected ozone depletion, especially for the high altitude layers. The method used here to correct for the aerosol effect still may have problems probably related to very small thickness of the layer. However considering the intrinsic variability of the ozone profiles and the other uncertainties related to the theoretical evaluation of the depletion effect, a large series of ozone data must be accumulated before any conclusion could be reached. The DIAL and aerosol lidar system we have described may be a simple and inexpensive way to accumulate these data with respect to other sampling technique (i.e. balloons) because the data acquisition rate may be limited only by weather conditions.

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