

Antarctic Ozone Transport and Depletion in Austral Spring 2002

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ABSTRACT

The ozone budget in the Antarctic region during the stratospheric warming in 2002 is studied, using ozone analyses from the Royal Netherlands Meteorological Institute (KNMI) ozone-transport and assimilation model called TM3DAM. The results show a strong poleward ozone mass flux during this event south of 45°S between about 20 and 40 hPa, which is about 5 times as large as the ozone flux in 2001 and 2000, and is dominated by eddy transport. Above 10 hPa, there exists a partially compensating equatorward ozone flux, which is dominated by the mean meridional circulation. During this event, not only the ozone column but also the ozone depletion rate in the Antarctic region, computed as a residual from the total ozone tendency and the ozone mass flux into this region, is large. The September–October integrated ozone depletion in 2002 is similar to that in 2000 and larger than that in 2001. Simulations for September 2002 with and without ozone assimilation and parameterized ozone chemistry indicate that the parameterized ozone chemistry alone is able to produce the evolution of the ozone layer in the Antarctic region in agreement with observations. A comparison of the ozone loss directly computed from the model's chemistry parameterization with the residual ozone loss in a simulation with parameterized chemistry but without ozone assimilation shows that the numerical error in the residual ozone loss is small.

1. Introduction

In September 2002, the Antarctic stratospheric vortex and its associated ozone hole elongated, displaced entirely from the pole, and subsequently split in two (e.g., Baldwin et al. 2003). As a result, a major stratospheric warming occurred (e.g., Varotsos 2002; Siegmund and van Velthoven 2004). Whereas over the Arctic major warmings occur about every other year (except in the 1990s, when they occurred in only two years), over the Antarctic only minor warmings and no major warmings have occurred since the beginning of the observational record in the late 1950s (Labitzke 1981; Labitzke and Naujokat 2000; Al-Ajmi et al. 1985). Despite its rarity, the breakdown of the vortex and the ozone hole was predicted accurately about a week in advance by the European Centre for Medium-Range Weather Forecasts (ECMWF) forecasting system (Simmons et al. 2005) and by the Royal Netherlands Meteorological Institute (KNMI) ozone-transport and assimilation model called TM3DAM (Eskes et al. 2005).

In the present study, the ozone budget in the Antarctic region during this event will be studied, using three-dimensional ozone analyses from the TM3DAM

model and wind analyses from ECMWF. For different polar caps, the ozone mass tendency inside and the ozone mass flux into the polar cap will be diagnosed for September–October 2002. The results will be compared with those for 2001 and 2000, when the ozone hole was quiescent as it is normally. Also the ozone depletion rate in the polar cap will be computed, as a residual of the ozone tendency and the ozone flux. To investigate the accuracy of this method, the residual ozone depletion rate will be compared with the ozone depletion directly computed from the model's chemistry parameterization. The zonal-mean meridional ozone mass flux will be investigated in detail, by separating it into the contributions by the mean meridional circulation and by eddy transport.

2. The TM3DAM model and data

TM3DAM is a three-dimensional ozone transport model with a parameterized stratospheric gas phase and heterogeneous ozone chemistry (Eskes et al. 2003). The gas phase production and loss of ozone is described by the linearized ozone chemistry parameterization developed and described by Cariolle and Déqué (1986), applying coefficients as provided by McLinden et al. (2000). It is used in diagnostic and in forecast mode. The diagnostic mode, applied in this study, is driven by archived 6-h forecast (ECMWF first guess) fields of wind, surface pressure, and temperature. The forecasts

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are updated every 6 h. The ozone analysis is based on total column ozone observations measured by the Global Ozone Monitoring Experiment (GOME) instrument onboard the *ERS-2* satellite of the European Space Agency (ESA; Valks et al. 2003). The model version used in this study produces an ozone analysis every 6 h and has 44 layers between 0.1 hPa and the surface and a horizontal resolution of $2.5^\circ \times 2.5^\circ$. The assimilation of GOME ozone observations and the forecast of ozone in TM3DAM are described in detail by Eskes et al. (2002, 2003). The present study applies TM3DAM ozone and ECMWF wind data for September–October 2002, 2001, and 2000.

3. Results

The transport of ozone into a polar cap around the South Pole is determined solely by the meridional ozone mass flux at the northern boundary of the polar cap. Therefore, the results for the ozone transport presented below will be focused on the meridional ozone flux (section 3a). The ozone budget in polar caps of different sizes will be considered in section 3b, and in section 3c, the effect of ozone assimilation and chemistry on the ozone budget and depletion in the polar cap will be investigated.

a. The meridional ozone mass flux

The meridional ozone mass flux f_{O_3} is defined as

$$f_{O_3} = O_3 v/g \quad (1)$$

where O_3 is the ozone mass mixing ratio, v is the meridional wind velocity, and g is the acceleration due to gravity. The f_{O_3} , O_3 , and v are functions of latitude, longitude, and altitude. The f_{O_3} is expressed in units of $\text{kg s}^{-1} \text{m}^{-1} \text{Pa}^{-1}$, that is, in kg s^{-1} per unit of “area” in the meridional plane (where the length units are m in the horizontal, and Pa in the vertical, i.e., pressure dimension). A positive value of f_{O_3} corresponds to a northward flux. The zonal mean of f_{O_3} can be written as

$$[f_{O_3}] = [O_3 v]/g = [O_3][v]/g + [O_3^*v^*]/g, \quad (2)$$

where $[x]$ and x^* denote, respectively, the zonal mean of x and the deviation of x from $[x]$. The two terms on the rhs of Eq. (2) represent the contributions to the zonal-mean meridional ozone mass flux by the mean meridional circulation and by eddy transport, respectively.

Figures 1a–c show the time versus pressure distributions of the zonal-mean meridional ozone mass flux at 60°S above 50 hPa in September and October 2002, 2001, and 2000, respectively. The latitude of 60°S has been chosen, because around this latitude the ozone fluxes during the event are largest (see below; Fig. 3a). Between about 21 and 28 September 2002, during the breakdown of the vortex, there exists a strong southward (poleward) ozone flux in the stratosphere be-

tween about 20 and 40 hPa, which is about 5 times larger than the flux during the remainder of September and October 2002 and during September–October 2001 and 2000. Between 21 and 28 September 2002, there also exists a strong compensating northward (equatorward) ozone flux above about 10 hPa. In the vertically integrated ozone mass flux (see below; Figs. 4b and 5b), the poleward flux dominates the equatorward flux. This is because the vertical extent of the poleward flux (about 20 hPa) is larger than that of the equatorward flux (about 10 hPa).

To investigate the strong zonal-mean meridional ozone mass fluxes at the end of September 2002 in more detail, Fig. 2 shows this flux (Fig. 2a) and the contributions to it by the mean meridional circulation (Fig. 2b) and by eddy transport [(Fig. 2c; following Eq. (2)]. Comparison of Figs. 2a–c reveals that the strong poleward ozone flux between 20 and 40 hPa is mainly due to eddy transport, whereas the strong equatorward flux at higher levels is mainly due to the mean meridional circulation. Around 10 hPa, the zonal-mean ozone flux is a small residual of large and opposite values of the contributions by the mean and eddy transport. The distributions of $[O_3]$ and $[v]/g$, which together determine the ozone flux by the mean meridional circulation (Fig. 2b), are shown in Figs. 2d and 2e, respectively. The pattern of the zonal-mean ozone flux closely resembles the pattern of $[v]/g$.

Figure 3 shows the same quantities as Fig. 2, but as a function of latitude, at 30 hPa. The level of 30 hPa has been chosen because around this level the ozone fluxes during the event are largest (see Fig. 2a). The largest poleward meridional ozone mass fluxes occur between about 55° and 80°S (Fig. 3a) and are dominated by eddies (Fig. 3c). As in Fig. 2, the pattern of the ozone flux by the mean meridional circulation (Fig. 3b) closely resembles the pattern of $[v]/g$ (Fig. 3e). The distribution of $[O_3]$ (Fig. 3d) clearly shows the large ozone increase during the event near the South Pole, from about 3×10^{-6} to $13 \times 10^{-6} \text{ kg kg}^{-1}$ between 21 and 26 September.

b. The ozone budget in the Antarctic region

The mass budget of ozone in a polar cap around the South Pole (the atmospheric volume between the South Pole and the northern boundary of the polar cap) can be expressed as

$$\frac{\partial M_{O_3}}{\partial t} = F_{O_3} + C_{O_3}, \quad (3)$$

where $\partial M_{O_3}/\partial t$ is the tendency of the ozone mass in the polar cap, F_{O_3} is the meridional ozone mass flux into the polar cap at its northern boundary, and C_{O_3} is the net chemical change of ozone in the polar cap. The F_{O_3} is computed as the integral of $-f_{O_3}$ over the meridional plane. The minus sign in the latter quantity has been included so that a positive value of F_{O_3} corresponds to a net positive ozone mass flux into the polar cap. The

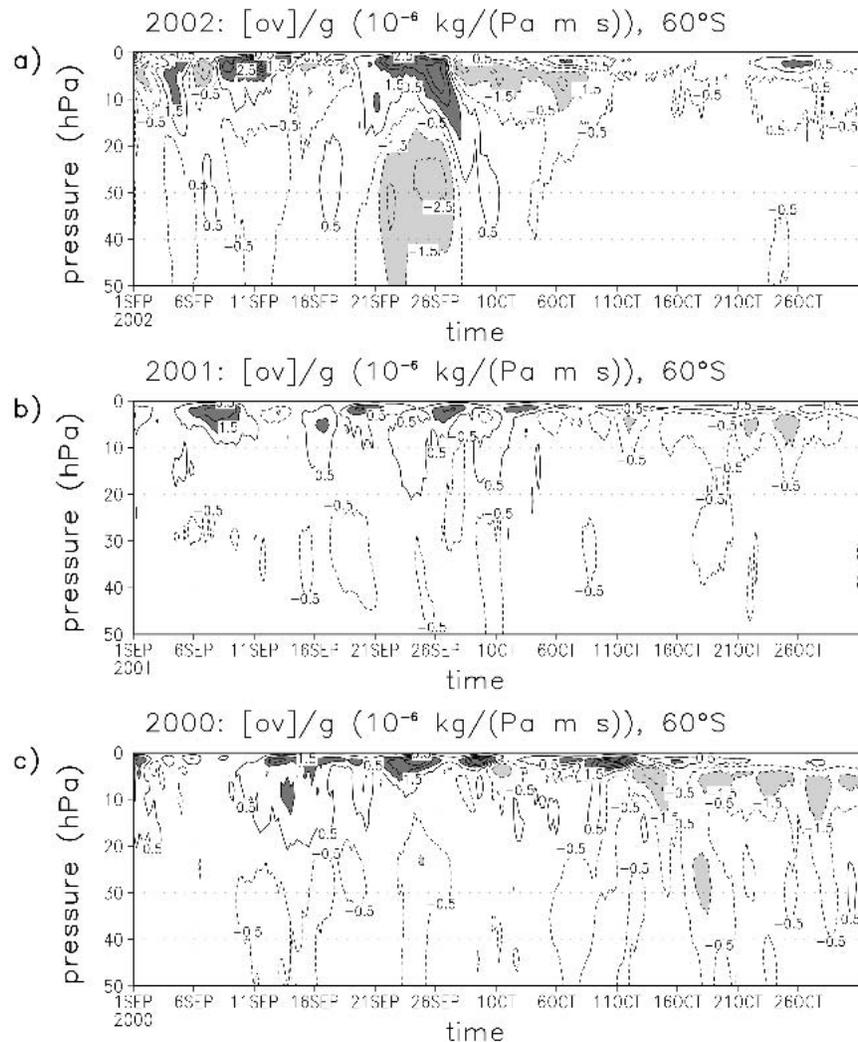


FIG. 1. Time vs pressure (hPa) distributions of the zonal-mean meridional ozone mass flux at 60°S in Sep–Oct of the year (a) 2002, (b) 2001, and (c) 2000. The contour values are ± 0.5 , 1.5 , 2.5 , $\dots \times 10^{-6} \text{ kg s}^{-1} \text{ m}^{-1} \text{ Pa}^{-1}$; dark shading: more than $1.5 \times 10^{-6} \text{ kg s}^{-1} \text{ m}^{-1} \text{ Pa}^{-1}$; and light shading: less than $-1.5 \times 10^{-6} \text{ kg s}^{-1} \text{ m}^{-1} \text{ Pa}^{-1}$.

quantities in Eq. (3) are expressed in units of kg s^{-1} . However, for convenience, the results for the quantities presented below will be expressed in Dobson units per day (DU day^{-1} , where $1 \text{ DU} = 2.89 \times 10^{16} \text{ molecules cm}^{-2}$). This conversion is obtained by first computing the quantities per unit of surface area of the polar cap [i.e., by dividing by $2\pi R^2(1 + \sin\phi_b)$, where R is the radius of the earth, and ϕ_b is the latitude of the northern boundary of the polar cap] and next by converting from $\text{kg s}^{-1} \text{ m}^{-2}$ to Dobson units (DU day^{-1} , where 1 DU day^{-1} corresponds to $2.48 \times 10^{-10} \text{ kg s}^{-1} \text{ m}^{-2}$). The $\partial M_{\text{O}_3}/\partial t$ and F_{O_3} are computed every 6 h from the TM3DAM ozone and ECMWF wind fields, and C_{O_3} is computed as a residual, applying Eq. (3). Here the $\partial M_{\text{O}_3}/\partial t$ is computed using centered differences around the times to which the F_{O_3} applies. The numerical error

of the residually computed net chemical change of ozone will be estimated in section 3c by comparing this residual with the net chemical change that is diagnosed directly from the model's chemical parameterizations.

Figure 4 shows several quantities relevant to the ozone budget in the polar cap south of 70°S in September and October of the years 2000, 2001, and 2002. After 20 September 2002, the average ozone column in this region (Fig. 4a; solid line) sharply rises from about 180 DU to more than 320 DU at 26 September, the latter value being more than 2 times larger than the values during this period in 2001 and 2000. After 26 September 2002, the ozone column gradually decreases to normal values in the middle of October and then gradually increases again, as in this period in 2001 and 2000. Both in the beginning and at the end of Septem-

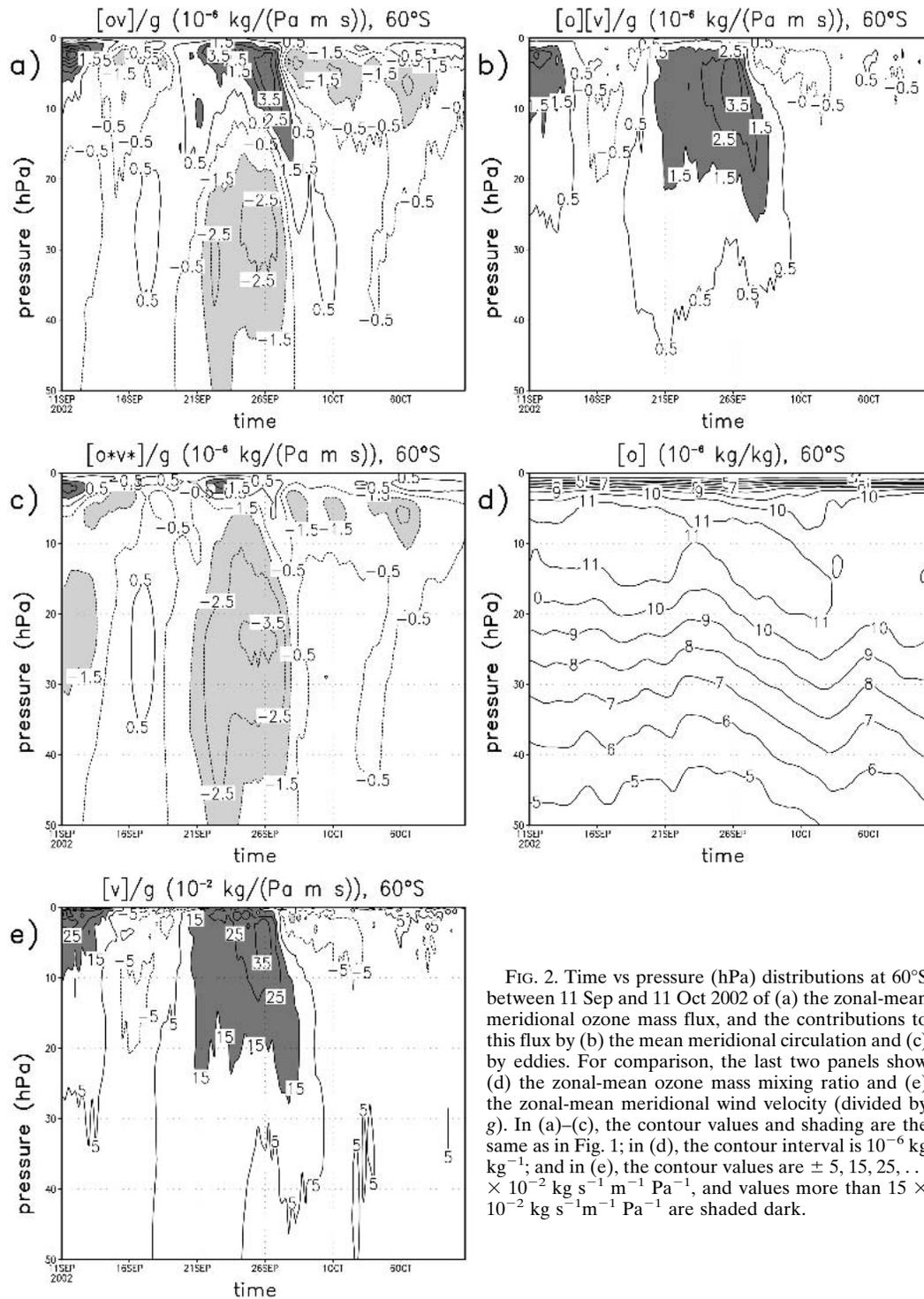


FIG. 2. Time vs pressure (hPa) distributions at 60°S between 11 Sep and 11 Oct 2002 of (a) the zonal-mean meridional ozone mass flux, and the contributions to this flux by (b) the mean meridional circulation and (c) by eddies. For comparison, the last two panels show (d) the zonal-mean ozone mass mixing ratio and (e) the zonal-mean meridional wind velocity (divided by g). In (a)–(c), the contour values and shading are the same as in Fig. 1; in (d), the contour interval is 10^{-6} kg kg^{-1} ; and in (e), the contour values are $\pm 5, 15, 25, \dots \times 10^{-2}$ $\text{kg s}^{-1} \text{m}^{-1} \text{Pa}^{-1}$, and values more than 15×10^{-2} $\text{kg s}^{-1} \text{m}^{-1} \text{Pa}^{-1}$ are shaded dark.

ber–October, the ozone column in 2002 is about 30% larger than in 2001 and 2000.

Figure 4b shows the ozone mass flux into the polar cap, which is identical to the meridional ozone mass flux at 70°S. After 20 September 2002, this flux first sharply increases from normal values to large positive

values of more than 50 DU day^{-1} at 22 September and next decreases back to normal values around 27 September. The ozone mass flux into the polar cap averaged over September–October is positive in each of the three years; its values are 9 DU day^{-1} in 2000 and 2002 and 6 DU day^{-1} in 2001.

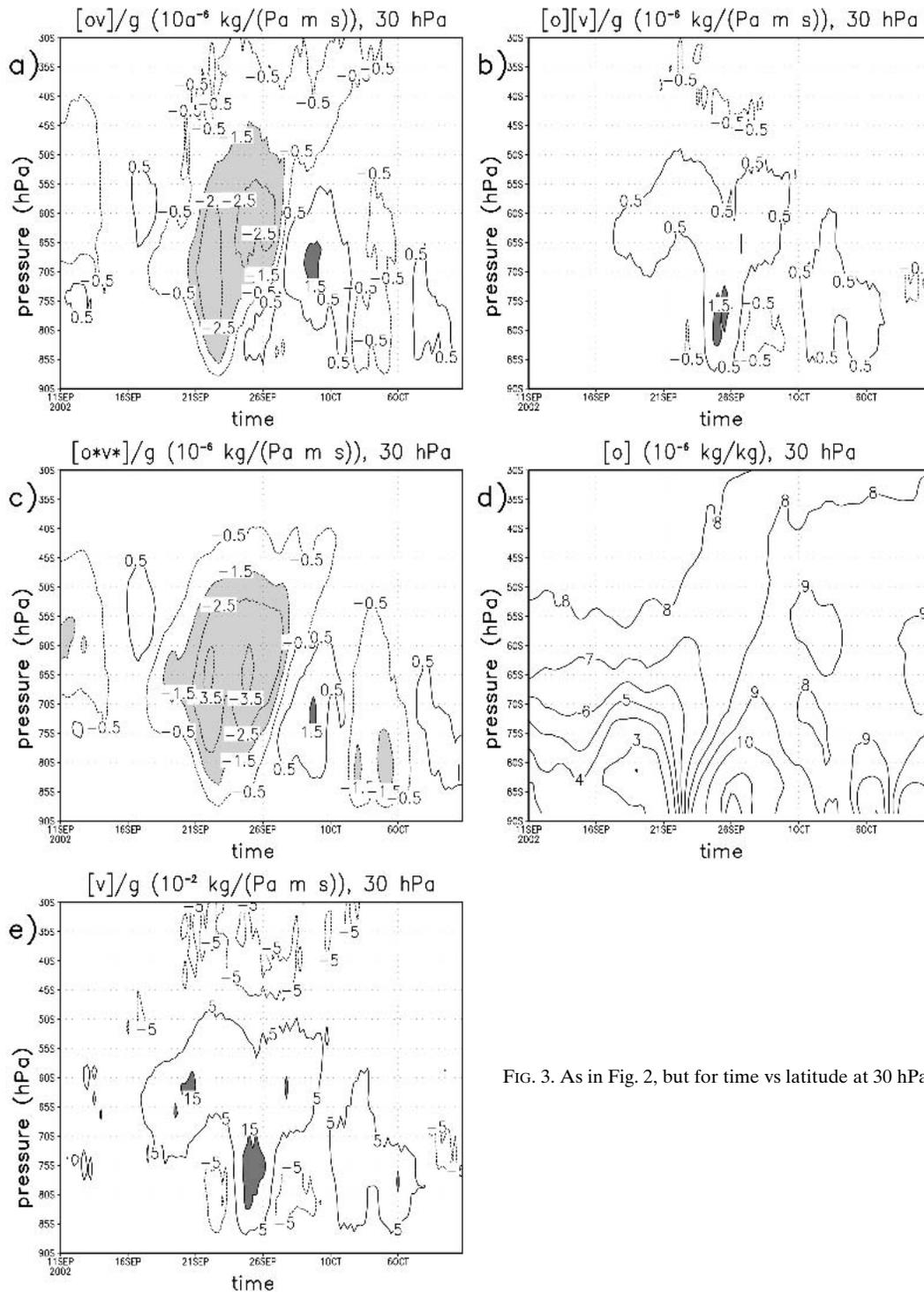


FIG. 3. As in Fig. 2, but for time vs latitude at 30 hPa.

Figure 4c shows the net chemical change during the entire period, both in 2002 and in the other two years. This term is computed as a residual from the assimilated ozone tendency in the polar cap and the flux of ozone into the polar cap, applying Eq. (3). Hereafter the term “chemical ozone loss” instead of “net chemical

change” will be used for situations where the net chemical change is negative. In 2002, the net chemical change in the first three weeks of September is somewhat larger (i.e., more negative) than in the other two years. In this period in 2002, the ozone hole was further formed, and the relatively large chemical change com-

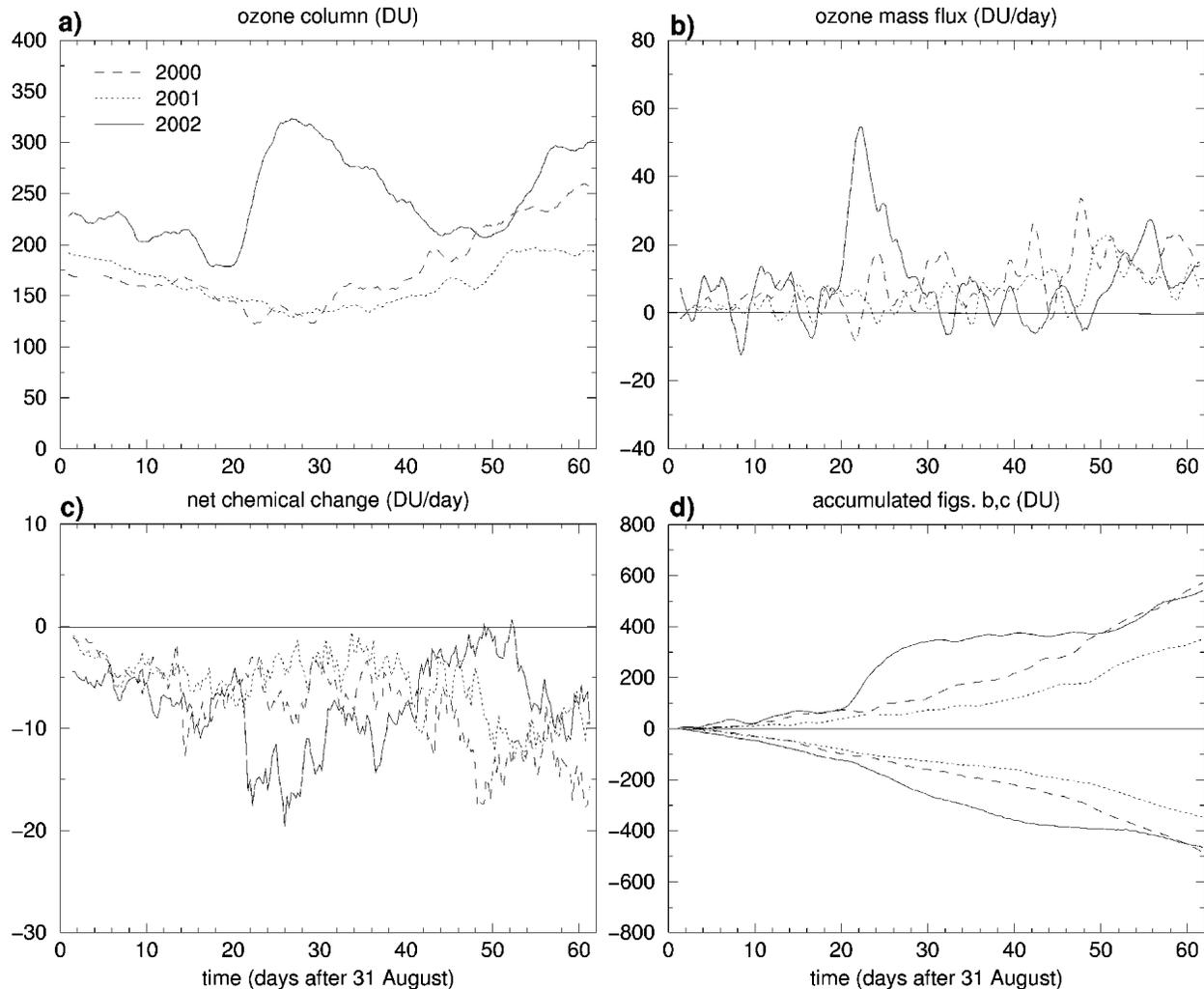


FIG. 4. Ozone budget quantities in the polar cap south of 70°S in Sep–Oct of the years 2000 (dashed lines), 2001 (dotted lines), and 2002 (solid lines): (a) the ozone column, (b) the ozone mass flux into the polar cap, (c) the net chemical change of ozone in the polar cap, and (d) the time integral of the ozone mass flux in (b) (positive values) and net chemical change in (c) (negative values). The quantities are expressed per unit of surface area of the polar cap in DU in (a) and (d) and DU day^{-1} in (b) and (c).

pensated for the rather slow start of the ozone hole in the weeks before September. Between about 22 and 30 September, the ozone loss rate is about 2 times larger than the loss during the remainder of the period. From an analysis of the parameterized ozone chemistry, it follows that this large ozone loss is mainly due to relaxation by this parameterization of the observed high ozone mixing ratio values toward the applied ozone climatology (not shown). As this large loss is also present in a simulation without ozone chemistry (see section 3c), it is probably not an artifact of the parameterization.

The accumulated ozone mass flux into and ozone loss in the polar cap during September–October, computed as the time integral of Figs. 4b,c, are shown by, respectively, the positive and negative curves in Fig. 4d. At the end of October, the accumulated ozone flux in 2002

is larger than in 2001, but, despite the very large ozone fluxes between 22 and 27 September 2002, it is not larger than in 2000. Similarly, the September–October accumulated ozone loss in 2002 is larger than in 2001 but does not strongly differ from the loss in 2000. This difference is small, because in 2000 the ozone loss in September–October is rather constant, whereas in 2002 it is large at the end of September but small in the middle of October (Fig. 4c). In 2001, the ozone hole existed for a longer period than in 2000 and 2002 (see Fig. 4a), and both the ozone mass flux into and the net chemical change in the polar cap were smaller than in 2000 and 2002. The accumulated ozone loss in September–October 2002 is about twice the average ozone column during this period, indicating a chemical time scale of about 1 month. As the bias in the residual ozone production depends on $\partial M_{\text{O}_3}/\partial t$ rather than on M_{O_3}

itself, its effect on the accumulated ozone production (Fig. 4d) is expected to be small.

The same quantities as shown in Fig. 4 are shown in Fig. 5, but for different polar caps in 2002, with northern boundaries at 60°, 70°, and 80°S. Whereas normally in September–October, the smallest ozone column values occur near the pole, Fig. 5a shows that at the end of September 2002, the largest ozone columns occur near the pole in the polar cap south of 80°S. Also, the ozone mass fluxes are largest for the polar cap south of 80°S (see Fig. 5b). This flux is largest—about 75 DU day⁻¹—on 22 September. The net chemical ozone loss on this day is much smaller—about 10 DU day⁻¹ (see Fig. 5c)—and, consequently, during this day the ozone column increases by about 65 DU (see Fig. 5a). In the first half of October, the average ozone column in the polar cap south of 80°S gradually halves from about 320 to 160 DU. Because in this period the ozone mass flux into the polar cap is very small (on average about 1 DU day⁻¹), this ozone column decrease is mainly due to chemical ozone loss. The accumulated ozone loss in September–October 2002 in the polar cap south of 80°S is about 2.3 times the average ozone column during this period, indicating a chemistry time scale of about 27 days. For the polar cap south 70°S, this time scale is about 32 days, and for the polar cap south of 60°S, it is about 50 days, indicating that the strength of the ozone depletion decreases with increasing distance to the pole. This variation in time scale is mainly due to the ozone tendency by gas phase chemistry. This tendency is negative poleward of about 45°S, and its magnitude increases toward the South Pole (see McLinden et al. 2000).

c. Simulations with and without ozone assimilation and chemistry

As mentioned in section 2, the ozone fields computed by the TM3DAM model are the result of the assimilation of GOME ozone observations in combination with parameterized ozone chemistry. In this subsection, the separate effects of assimilation and chemistry on the ozone column and the net chemical change of ozone will be investigated. This will be done by analyzing four simulations with the TM3DAM model for September 2002: 1) with assimilation, with ozone chemistry (hereafter ac); 2) no assimilation, with chemistry (nac); 3) with assimilation, no chemistry (anc); and 4) no assimilation, no chemistry (nanc). For all four simulations, the same initial ozone conditions have been applied.

The ozone column and the residually computed net chemical change in the polar cap south of 70°S in these four simulations are shown in Figs. 6a and 6b, respectively. The evolution of the ozone column in the nac simulation (thin dashed line) is nearly the same as that in the ac simulation (thick solid line). Thus, the parameterized chemistry alone is able to produce the evolution of the ozone column in agreement with the GOME observations. There are, however, striking differences

between the nac and the ac simulation that particularly become visible in the time derivative of the ozone column, from which the net chemical change is derived. As is shown in Fig. 6b, at the end of September 2002, the residual net chemical change in the nac simulation is much smaller (i.e., less negative) than that in the ac simulation.

Before attempting to explain this difference, we first consider the fact that the net chemical change that is residually computed from the ozone tendency and the ozone mass flux, using Eq. (3), is inherently prone to numerical errors. We estimate these errors by also deriving the net chemical change directly from the chemistry parameterization in the TM3DAM model and by then comparing the result with the residual net chemical change in the nac simulation. Since in the nac simulation the chemical change is entirely due to the parameterized chemistry, ideally the residual net chemical change computed from this simulation should be equal to the directly computed net chemical change, and their difference indicates the numerical error in the residually computed net chemical change. These residually and directly computed net chemical changes are shown by, respectively, the dashed and solid lines in Fig. 7 (the residual result is also shown by the dashed line in Fig. 6b). On a daily basis, the difference between the two results is, on average, about 8%. A possible explanation for this difference is that the ozone column tendency at time t_0 , from which the residual net chemical change at t_0 is determined, is computed as a centered difference of the ozone columns at $t_0 + 6$ h and $t_0 - 6$ h, whereas the direct net chemical change is determined from data that all apply to t_0 . The difference between the two monthly averaged results is less than 1%. Thus, the systematic difference between the residually and the directly derived net chemical change is very small.

As the difference in the residual chemical change between the ac and the nac simulation is much larger than 8%, this difference cannot be explained by numerical errors alone. It must be either due to errors in the parameterized chemistry, which in the ac simulation are corrected by the assimilation, or by errors in the assimilation, which are absent in the nac simulation, or both. There are reasons to assume that the large chemical change at the end of September 2002 in the ac simulation is too large, due to a bias in the GOME ozone observations. A comparison of GOME with ground-based observations of total ozone [M_{O_3} in Eq. (3)] shows that in September–October in the South Polar region, the bias in M_{O_3} (GOME minus ground-based) decreases with increasing value of M_{O_3} and increasing solar zenith angle (P. Valks 2004, personal communication). The latter effect leads to an erroneous decrease with time of the observed ozone column and, hence, to an overestimation of the net chemical change (i.e., the residual C_{O_3} will be too negative). With respect to the former effect, when $\partial M_{O_3}/\partial t$ is positive, it will be underestimated by the GOME measurements, and the re-

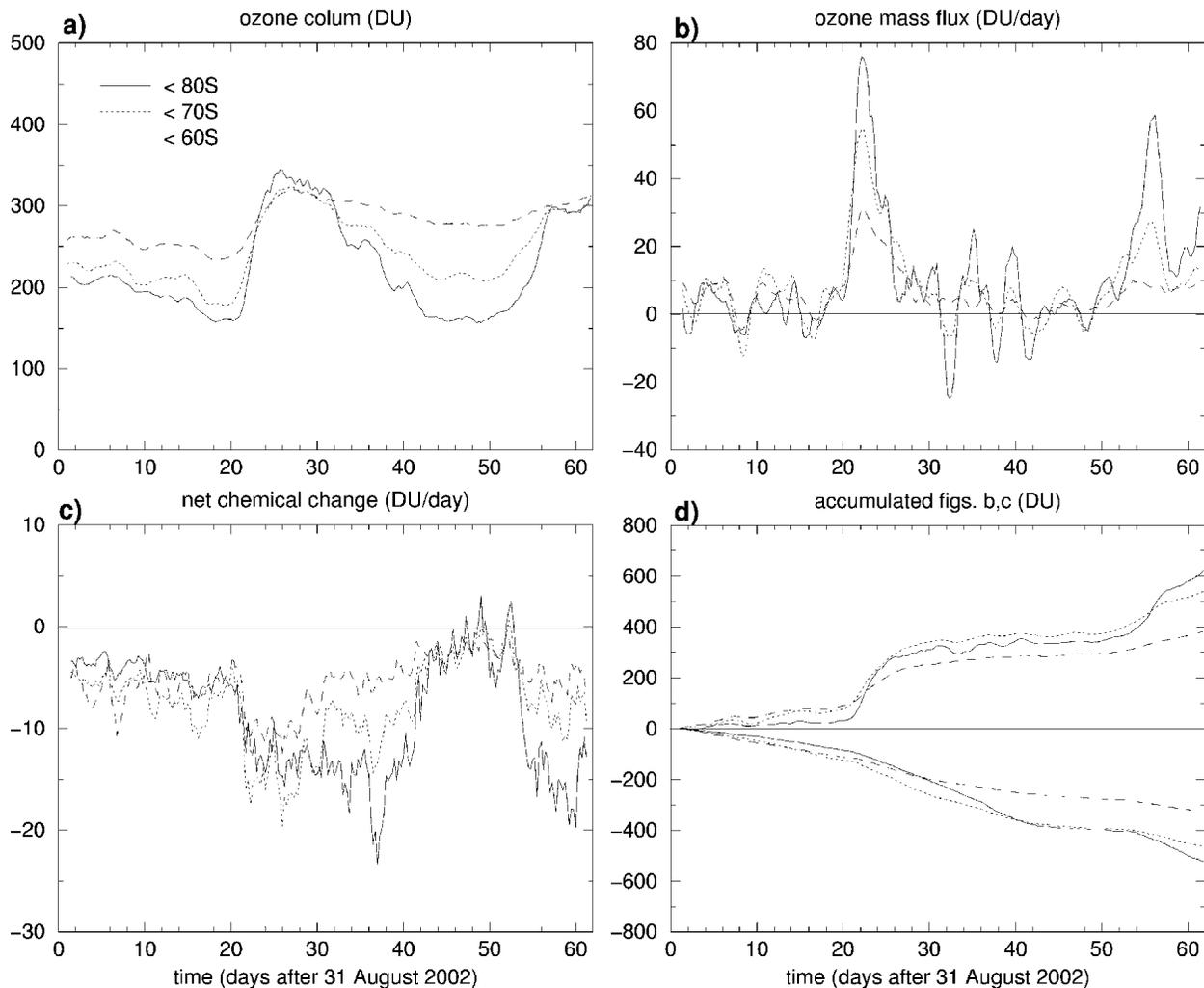


FIG. 5. As in Fig. 4, but for polar caps of different sizes, with northern boundaries at 60° (dashed lines), 70° (dotted lines), and 80°S (solid lines) in Sep–Oct 2002.

sidual net chemical change will be overestimated. As between about 20 and 25 September 2002, the $\partial M_{O_3}/\partial t$ is strongly positive; it is expected that the residual ozone loss in the ac simulation in this period is too large. Although the factors determining the bias in the GOME observations are known qualitatively, they are poorly known quantitatively, and, hence, the bias in the computed residual net chemical change can presently not be accurately determined.

In the nanc simulation, there is no chemical ozone loss, either from the assimilation of GOME observations or from the parameterized ozone chemistry. Therefore, the ozone column values in this simulation (Fig. 6a; thick dashed line) are larger than in the ac simulation. At the end of September, the difference is about 80 DU. This difference, however, is much smaller than the accumulated ozone loss in the ac simulation at the end of September (about 260 DU; see Fig. 4d). Therefore, the ozone mass flux into the polar cap in the

nanc simulation must be about $260 - 80 = 180$ DU smaller than in the ac simulation. Thus, the absence of ozone assimilation and ozone chemistry leads to a smaller ozone flux into the polar cap, indicating that outside the polar cap there is a net positive production of ozone. As in the nanc simulation, there is no net chemical change; in the absence of numerical errors the residual net chemical change should be zero. As is shown in Fig. 6b (thick dashed line), this residual is indeed almost zero, indicating, again, that the numerical errors in its computation are very small. This conclusion was also obtained above from the results in Fig. 7.

To investigate the ability of assimilation alone to capture the evolution of the ozone columns, we performed a simulation with assimilation of GOME observations and without parameterized ozone chemistry (the anc simulation). In the beginning of September, the net chemical change in the anc simulation is much smaller

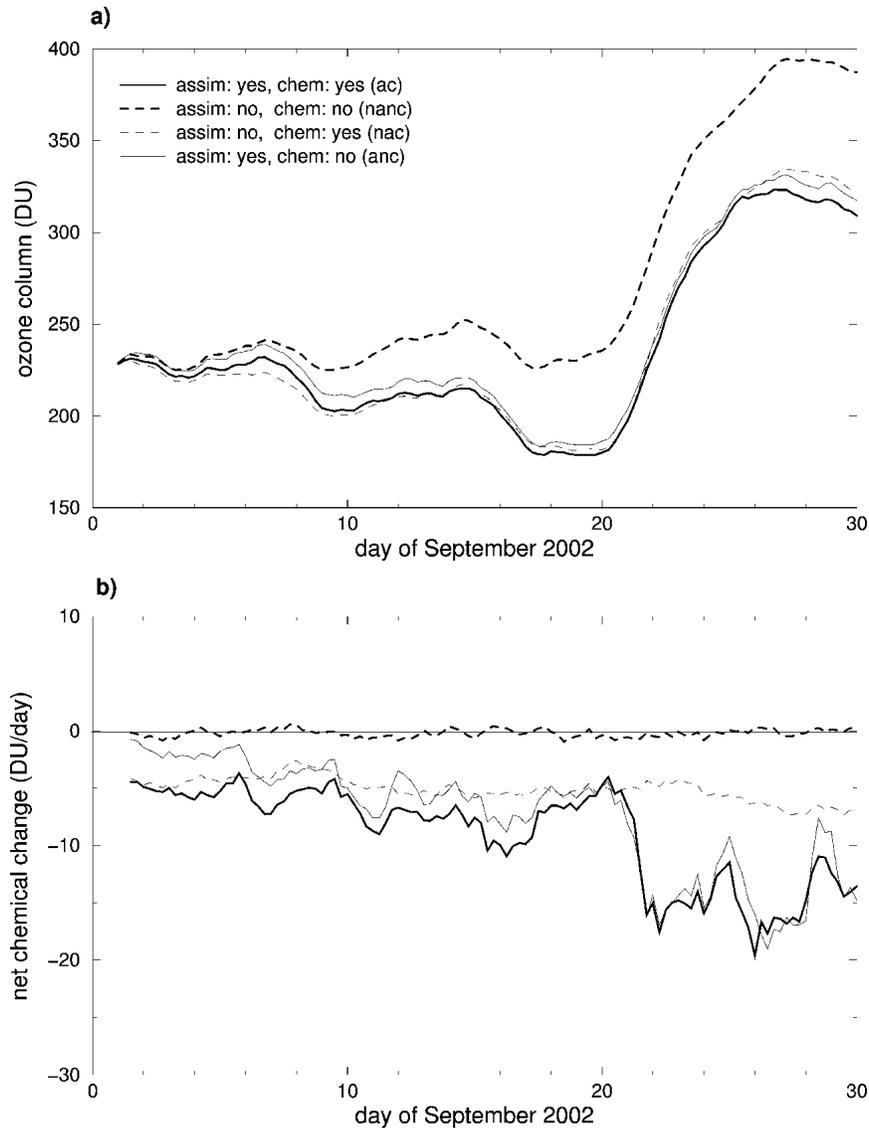


FIG. 6. (a) Ozone column (in DU) and (b) residually computed net chemical change of ozone (in DU day^{-1}) in the polar cap south of 70°S in Sep 2002 in four simulations with or without ozone assimilation and chemistry: ac simulation (thick solid lines), nanc simulation (thick dashed lines), nac simulation (thin solid lines), and nac simulation (thin dashed lines).

than in the ac and nac simulations. This is because in this period the polar region, where most of the chemical change occurs, is dark and, therefore, poorly observed by the GOME instrument. During the course of September, the polar night gradually disappears, the coverage by GOME of the polar region improves, and the net chemical change approaches the values that are obtained with the ac simulation in which the ozone chemistry is explicitly parameterized. Also, the ozone columns in the anc simulation are close to those in the ac simulation, implying that the ozone columns in the ac simulation are mainly determined by the assimilation of GOME observations and that the parameterized chem-

istry only plays a minor role. This also means that possible shortcomings of the parameterized ozone chemistry have only little effect on the assimilated ozone column and on the residually computed net chemical change of ozone.

4. Conclusions

In conclusion, the results show that the sharp total ozone increase after 20 September 2002 is mainly due to a large poleward ozone mass flux between about 20 and 40 hPa, associated with strong eddy activity. The

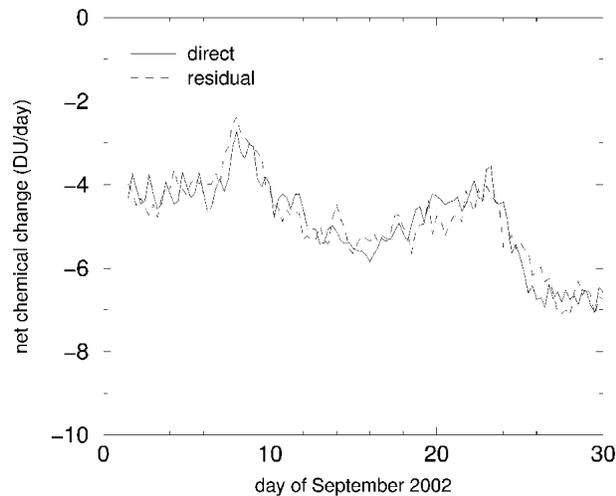


FIG. 7. The net chemical change of ozone in the polar cap south of 70°S in Sep 2002 computed directly from the chemistry parameterization in the TM3DAM model (solid line) and computed as a residual from the ozone column tendency and ozone mass flux in the nac simulation (i.e., without assimilation, but with parameterized ozone chemistry) (dashed line). Units are DU day⁻¹.

gradual ozone decrease in the first half of October 2002, on the other hand, is mainly due to ozone depletion, whereas the final ozone increase at the end of October 2002 is again mainly due to poleward transport of ozone. Also, in September–October 2000 and 2001, a poleward transport of ozone occurred, approximately balancing the ozone depletion, but its amplitude was about 5 times smaller than at the end of September 2002. Despite the high temperatures in the polar vortex in September 2002 (Baldwin et al. 2003), the chemical ozone loss during September–October 2002 is comparable to that in the same period in 2000, when the ozone hole was extremely large (Canziani 2001).

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