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Global Fields of the Total Ozone and Nitrogen Dioxide Contents Retrieved from Satellite Measurements and a Three-Dimensional Simulation

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Abstract—The global fields of the total ozone and nitrogen dioxide contents retrieved from satellite measurements and from simulations based on a transport–photochemical atmospheric model are compared to each other. For the comparison, the daily GOME (Global Ozone Monitoring Experiment) satellite global data monthly averaged over January, April, July, and October 1998 and the data obtained for analogous conditions by simulation with the MEZON model are used. The main features of the spatial distributions of total ozone and nitrogen dioxide observed in the data of GOME satellite measurements, namely, the characteristic latitudinal distributions and local regions of increased and decreased concentrations, are reproduced in MEZON simulations. Significant (by several times) discrepancies between the NO_2 total contents obtained from satellite measurements and simulations are revealed for central Europe, eastern areas of the United States, China, and other regions characterized by increased anthropogenic pollution of the troposphere.

INTRODUCTION

Over the last several decades, significant progress was achieved in the understanding of the features of the space–time distribution of atmospheric minor gas constituents (MGCs). This progress was promoted by modernization of global satellite and ground-based measuring systems and by a significant development of three-dimensional models for atmospheric photochemistry and dynamics. The use of real meteorological fields from data assimilation systems [1–3] allows the reproduction of MGC distributions with high spatial and temporal resolutions and offers the possibility of a detailed comparison between the atmospheric gas compositions simulated and directly or indirectly measured. Such a comparison enables us to assess the degree of our understanding of atmospheric processes and to determine which components of the model should be improved first. On the other hand, it is well known that a priori information taken from different models is frequently used to solve the problem of MGC concentration retrieval from remote measurements. In this connection, as detailed an intercomparison as possible of the results of remote sounding performed by different methods and from different platforms with one another and with simulated results, which reflect the accumulated knowledge on atmospheric processes, is of importance. This intercomparison will make it possible, on the one hand, to reduce uncertainty in the data of indirect sounding and, on the other hand, to reveal the gaps in our understanding of chemical and dynamic atmospheric processes.

In this work, we compare the global distributions of the total ozone and nitrogen dioxide contents retrieved from the GOME (Global Ozone Monitoring Experiment) satellite measurements and computed using the MEZON three-dimensional global model. This is the first comparison between the field of the total NO_2 contents simulated by the MEZON model and that retrieved from satellite measurements. Previously, different authors performed similar comparisons between simulations and GOME measurements [4, 5]. However, they studied the NO_2 distributions in the troposphere only, which requires a special interpretation for the GOME data and, in our opinion, introduces an additional uncertainty into the results of comparisons. In Section 1, the GOME satellite experiment is presented. The MEZON model is considered in Section 2. Section 3 is devoted to a comparison between the fields of the total ozone and NO_2 contents obtained from GOME measurements and MEZON simulations. Section 4 presents the principal results and conclusions of this work.

1. SATELLITE MEASUREMENTS

The GOME instrument is one of the instruments installed aboard the ERS-2 satellite launched in April 1995 [6]. The GOME instrument performs nadir measurements of the outgoing reflected and scattered radiation in the UV and visible spectral regions (240–790 nm) with a nominal spatial resolution of 40 × 320 km and allows virtually global monitoring of the total ozone and NO_2 content distributions.

As was shown in many works (see, for example, [7]), the potential accuracy of the total ozone and NO_2 content measurements with the GOME instrument is rather high, namely, the errors are estimated at less than 1%. It is important to note that such estimates characterize the maximum accuracy of the measurements under consideration. The errors assigned to the GOME data are estimated at about 1 and 10% for the total contents of ozone and NO_2, respectively. The actual retrieval errors are significantly greater because of a large number of factors usually disregarded in estimations of the potential accuracy. Each satellite experiment includes specialized studies intended to estimate the actual accuracy of measurements and the characteristics of retrieved data. In this connection, from the outset of the GOME instrument's operation, a large number of comparisons of its measurements with those performed by other space- and ground-based measuring systems were performed [8].

A comprehensive review of international experiments on the validation of the total ozone content (TOC) measurements performed with the GOME instrument is presented in [9]. Among other things, this review shows that a large number of comparisons between the GOME routine data on the TOC and the data obtained independently with the ground-based instruments of the Russian ozonometric network in 1996–2000 indicates that the TOC values measured with the GOME instrument are systematically underestimated by 4 to 9%. A comparison of the simultaneous data on the TOC values obtained with the GOME instrument and ground-based instruments of seven Russian stations shows that the yearly mean systematic underestimation of the TOC values in the GOME data is about 4% [10].

A number of authors compared the GOME data on the total NO_2 content with the data of ground-based measurements, which are less numerous than TOC measurements, and demonstrated that the satellite and ground-based data are in agreement within their total error (< 20%) [11]. Meanwhile, a significant discrepancy was revealed between the GOME and ground-based data obtained under high tropospheric pollution. For example, a comparison between the data on the total NO_2 content obtained with the GOME instrument and the ground-based instrument installed at Zvenigorod (Russia) [12] shows that the rms discrepancies are about 70%. Most similar comparisons indicate that the temporal changes in the NO_2 content are stepwise and the values of the total NO_2 content obtained with the GOME instrument exceed those obtained from the ground-based measurements. The latter result is usually attributed to the different sensitivities of the satellite and ground-based measuring systems to the anthropogenic pollution of the lower troposphere.

Table 1. Comparison of the TOC global fields retrieved from the GOME measurements performed in 1998 with those simulated (mean value, rms discrepancy, and correlation coefficient)

Month	$\Delta_{\text{GOME-MEZON}}$, %	$\Delta_{\text{GOME-MEZON}}$, %	R
January	–2	6	0.94
April	–1	10	0.92
July	–7	11	0.91
October	–11	15	0.86

2. GLOBAL THREE-DIMENSIONAL TRANSPORT–PHOTOCHEMICAL MODEL MEZON

MEZON (Model for Investigating Ozone Trends) is a global model of transport and photochemical atmospheric processes. It has horizontal latitudinal and longitudinal resolutions of 4° and 5°, respectively. The model atmosphere extends in the vertical direction from the Earth's surface to a level of 1 hPa and is divided into 24 layers. It consists of four principal components: photochemical, transport, and radiation blocks and a database including daily values of the temperature and wind velocity [13].

The photochemical block of the model allows the computation of the concentrations of 43 atmospheric gases, which are controlled by 199 gaseous-phase and 16 heterogeneous reactions. The photolysis rates should be computed at each step of integration by using the current values of the ozone mixing ratio and solar radiation flux and the procedure of tabular interpolation. The chemical transformations and transport of MGCs are computed using a two-hour step.

To construct the wind and temperature fields, the MEZON model assimilates the UKMO (UK Meteorological Office, http://www.meto.govt.uk/) databases [14]. For the initial conditions corresponding to September 1992, the concentrations of O_3, CH_4, NO_x, and other MGCs are taken from the UARS (Upper Atmosphere Research Satellite, http://hyperion.gsfc.nasa.gov) climatological database. For each integration year, the surface concentrations of CO_2, CH_4, and N_2O, etc., are taken from the NOAA/CMDL (http://www.cmdl.noaa.gov) database separately for the Southern and Northern hemispheres. The surface fluxes of NO_x and CO are taken from the NCAR (National Center for Atmospheric Research, http://www.ncar.ucar.edu/ncar/) database.

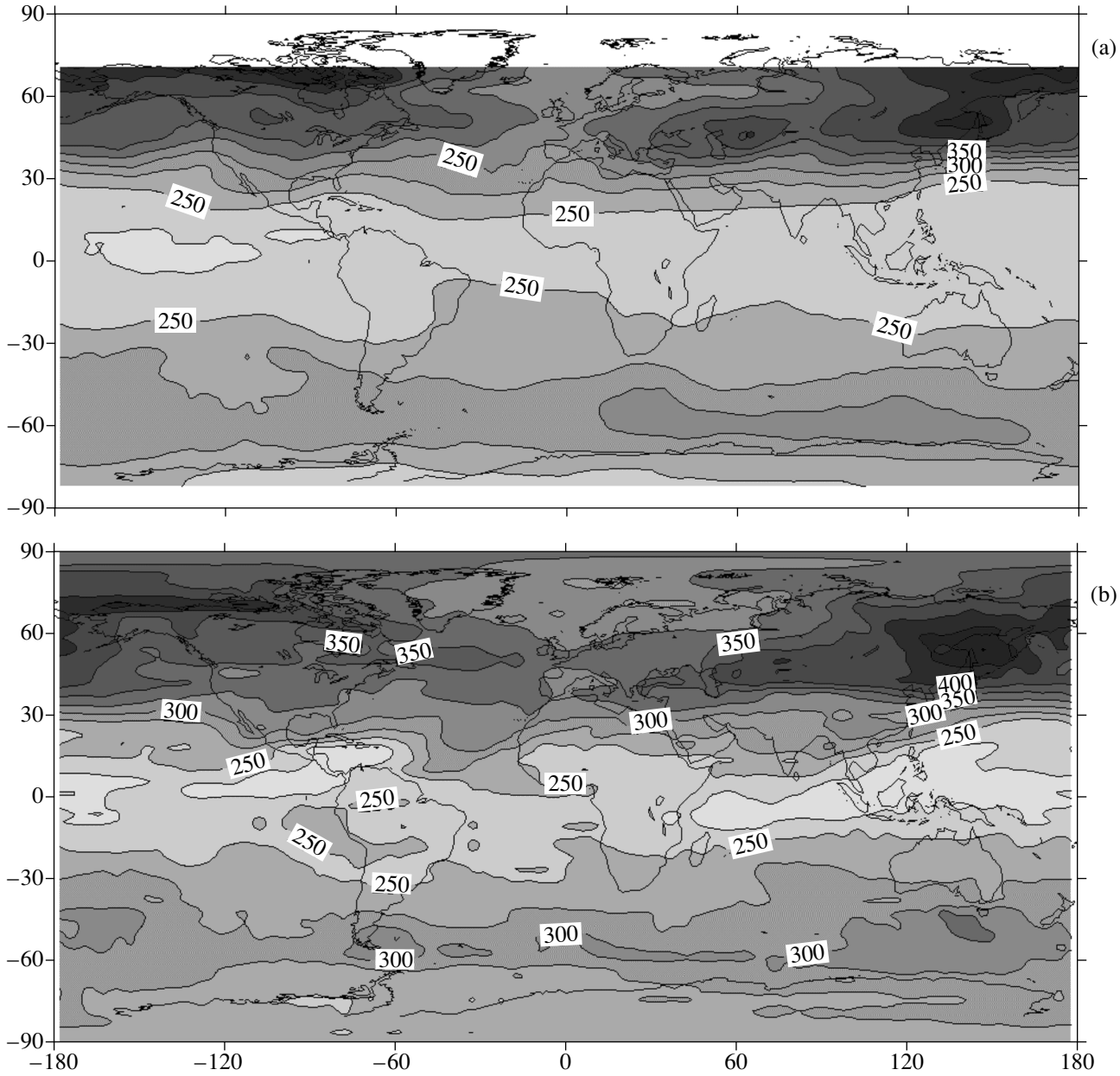

Fig. 1. Global field of the total ozone content (DU) obtained from (a) measurements and (b) simulation; January 1998.

The MEZON model is described in more detail in [3, 13, 15]. The zonal mean distributions of the ozone and NO_x mixing ratios were validated in [15] by comparing between the MEZON data and the UARS database. A comparison of the TOC model geographic distribution with the TOMS database was performed for each March over the period 1993–2000 [16]. The results indicated that the MEZON model satisfactorily reproduces the MGC distributions and the TOC interannual variability observed in March over the Northern Hemisphere.

In this work, the model fields of the total ozone and NO_2 contents computed for a four-month period of 1998 are validated on the basis of their comparison with the GOME data. To determine the monthly mean ozone and NO_2 fields needed for the comparison with the GOME data, the MGC distributions were computed from the 1998 UKMO meteorological fields with the total ozone and NO_2 contents recorded at local noon, which corresponds to the time of the GOME measurements.

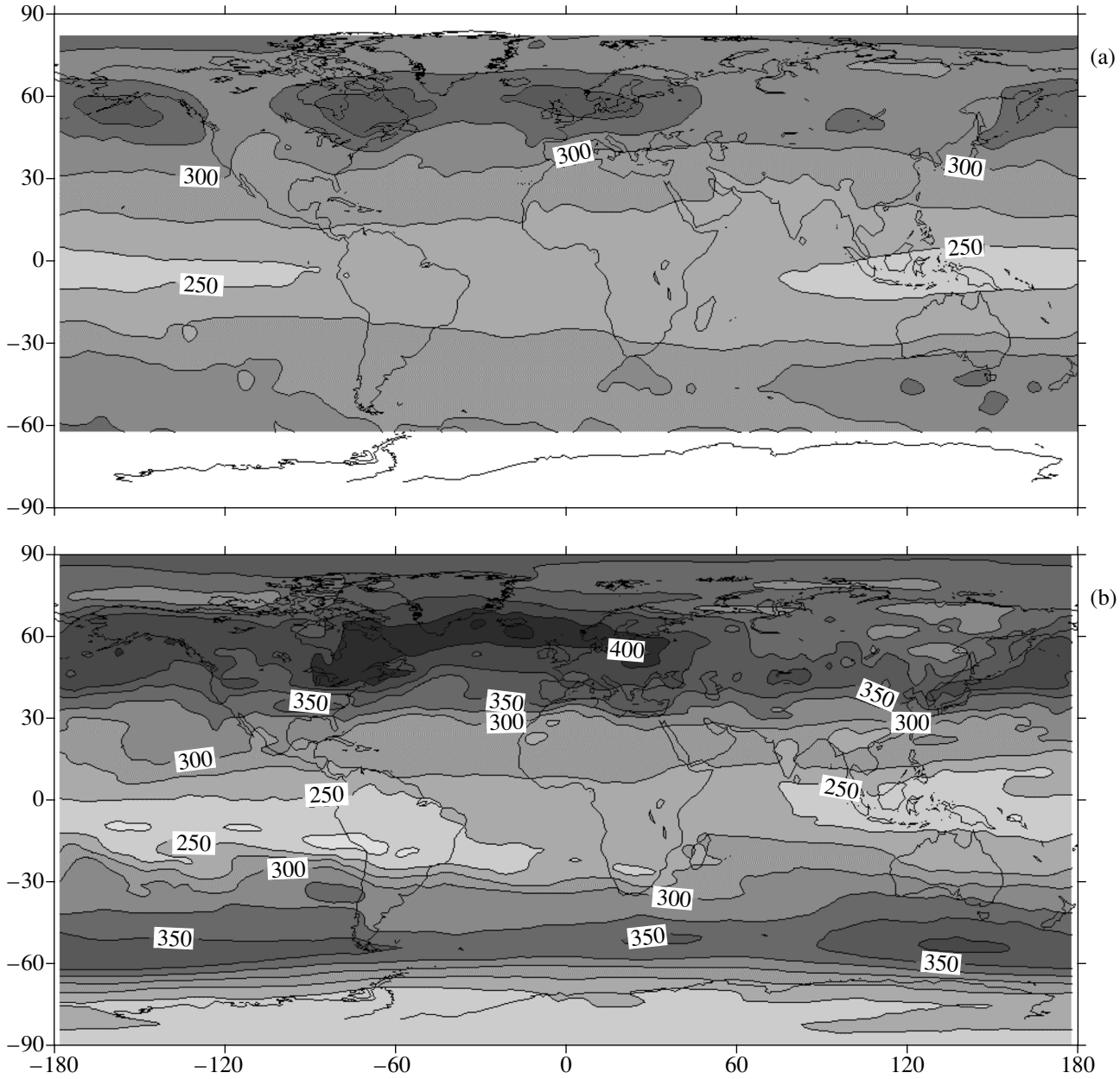

Fig. 2. Global field of the total ozone content (DU) obtained from (a) measurements and (b) simulation; July 1998.

3. TOTAL OZONE AND NO_2 SPATIAL DISTRIBUTIONS BASED ON GOME MEASUREMENTS AND MEZON SIMULATIONS

For a comparative analysis of the measured and simulated spatial distributions of total ozone and NO_2, we considered the global fields of the monthly mean total contents of these constituents for four months (January, April, July, and October) corresponding to four different seasons. The satellite data necessary for this comparison with the model were represented in the form of global fields obtained by the authors as a result of averaging all GOME measurements performed for one month. The results of GOME measurements performed in 1998 were used. The region of each GOME measurement (a rectangle 40×320 km, or a scan) was projected onto a global grid with a step of 40 km. At the points of scan overlapping, the measured values were averaged. Then, the resulting field characterized by a horizontal resolution of 40 km (about 0.3°) was transformed to a grid corresponding to model spatial latitudinal and longitudinal resolutions of 4° and 5°, respectively. These fields were compared with analogous fields computed on the basis of the MEZON model for local noon, at which the GOME measurements are performed. The results of such comparisons are given in the form of maps of relative discrepancies (GOME value minus MEZON value) and in the form of tables listing the mean values, rms discrepancies, and correlation coefficients.

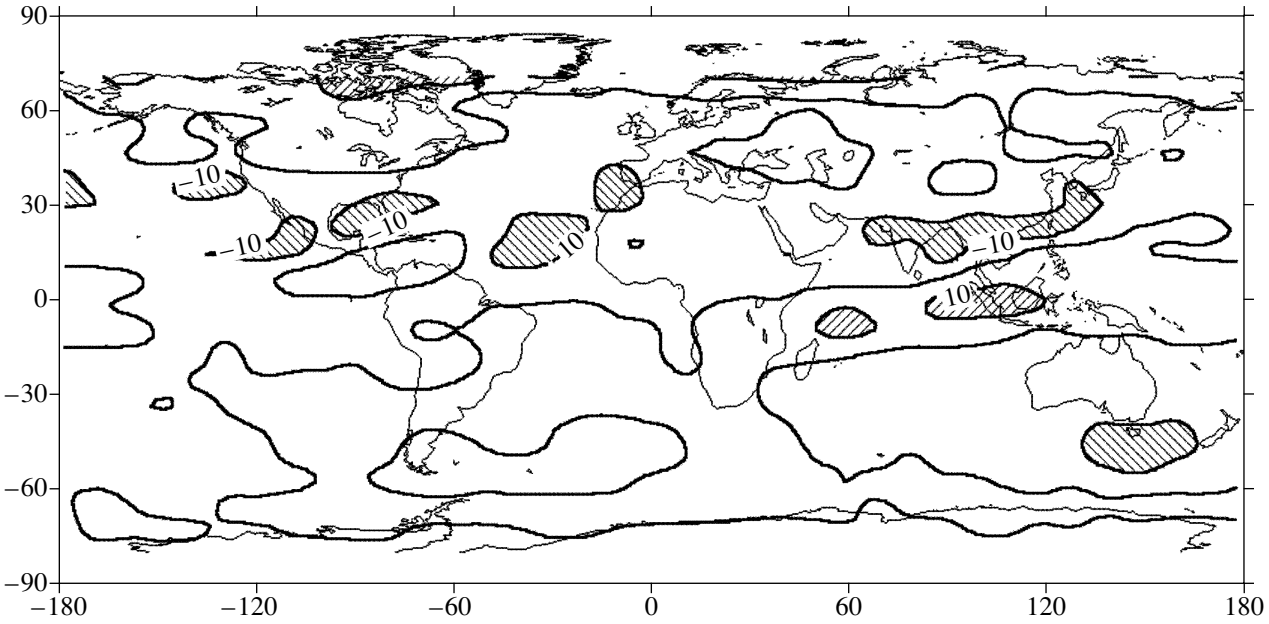

Fig. 3. Map of relative discrepancy between data on the total ozone content (%): (GOME–MEZON)/GOME; January 1998.

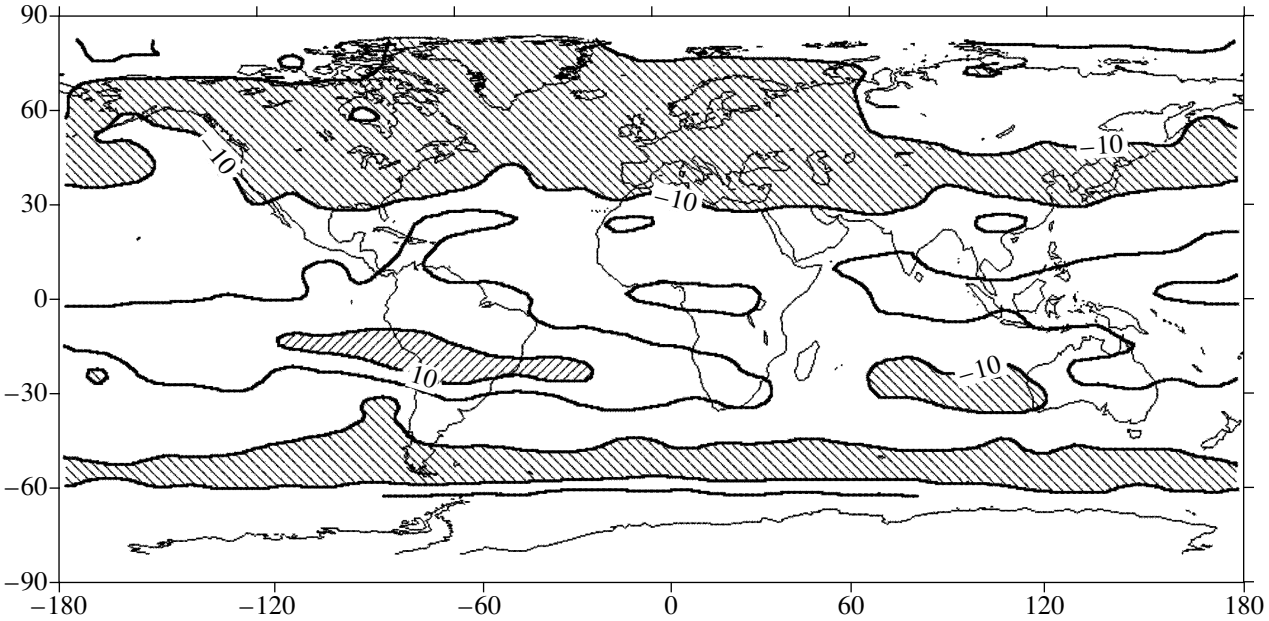

Fig. 4. Map of relative discrepancy between data on the total ozone content (%): (GOME–MEZON)/GOME; July 1998.

3.1. Global Distribution of the Total Ozone Content

The TOC spatial distributions computed for the months under consideration are characterized by April maxima (more than 450 DU) localized over Alaska, Canada, and Siberia. These maxima are caused by the well-known winter–spring processes of ozone accumulation in these regions. Over the Antarctic region, an October minimum (less than 150 DU), which is associated with the phenomenon of the so-called ozone hole, is observed. The results of comparisons between the TOC global fields computed on the basis of MEZON simulations and GOME measurements are presented in Figs. 1 and 2 for January and July, respectively. On the whole, the model well describes the principal features of the TOC spatial distribution observed in these seasons, namely, it reproduces the tropical belt of a depressed ozone content and the regions of a relatively high TOC localized in the middle latitudes of both hemispheres. For example, the GOME measurements

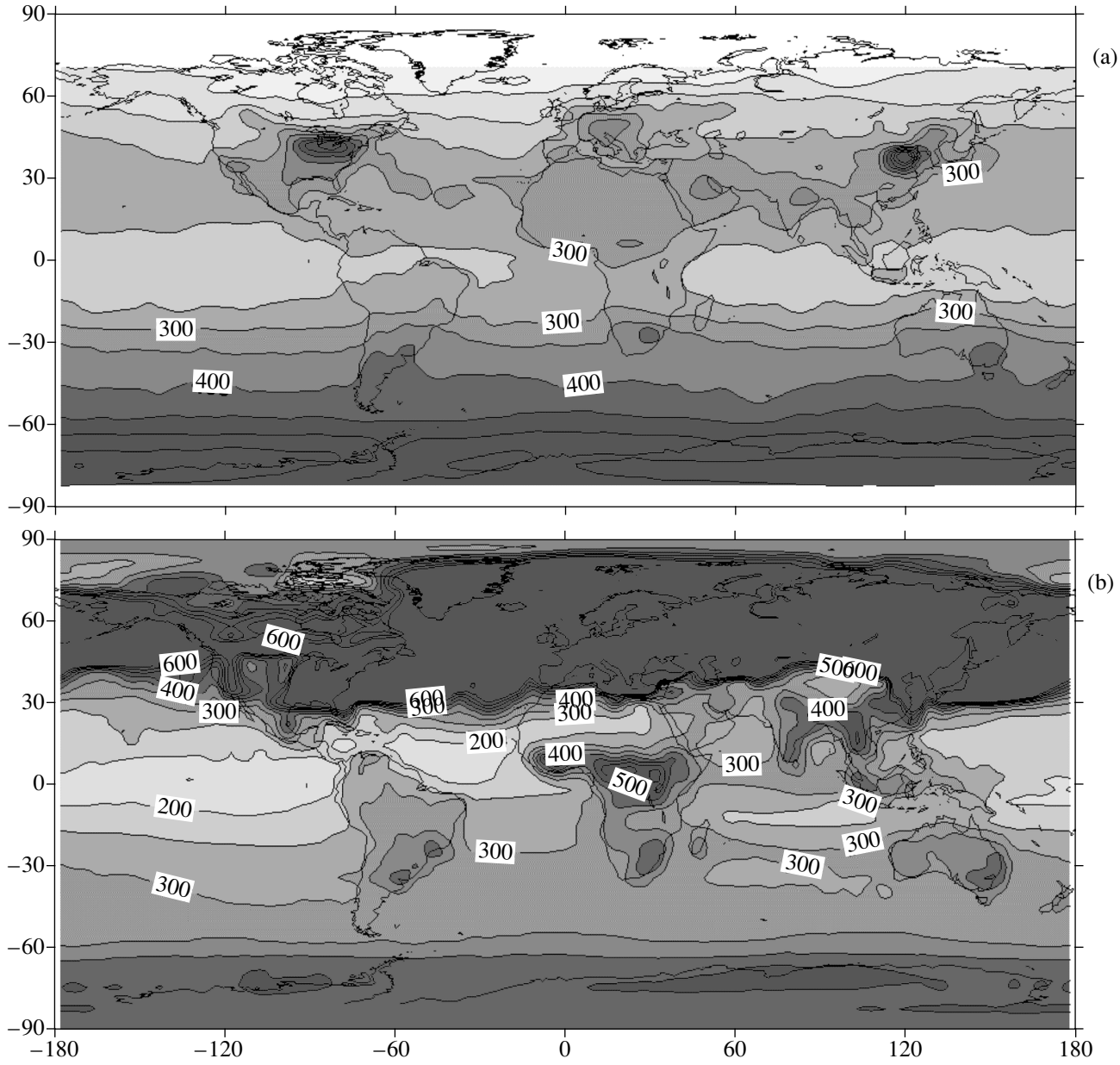

Fig. 5. Global field of the total NO_2 content (10^{13} mol/cm^2) obtained from (a) measurements and (b) simulation; January 1998.

show that clearly defined regions of increased TOC occur in January over Siberia (350–400 DU), the Sea of Okhotsk (about 450 DU), and Alaska and northern Canada (400–450 DU). This result correlates well with the simulated results (see Fig. 1). In the GOME TOC field, a region of relatively high ozone content (about 350 DU) can be noted for July over eastern Canada. This phenomenon is also predicted by the model (see Fig. 2). For January and July, the occurrence of several regions of increased TOCs in the middle latitudes of the Southern Hemisphere is revealed from both satellite measurements and simulations.

The estimated statistical characteristics of the results of the comparison, namely, the mean and rms discrepancies (Δ and σ, %) between the data obtained with the GOME and MEZON procedures and the correlation coefficient R are presented in Table 1. The best agreement between the measured and simulated data is observed for January and April: $\Delta = -2$ and –1%, $\sigma = 6$ and 10%, and $R = 0.94$ and 0.92, respectively. The worst agreement between the GOME and MEZON data is observed for October: $\sigma = 15\%$ and $R = 0.86$. It can be noted that the results based on the model simulations always exceed the results based on the GOME measurements by about 10% on the average. However, it is necessary to note that the validation of the GOME TOC data (Section 1) shows that the GOME TOC values are

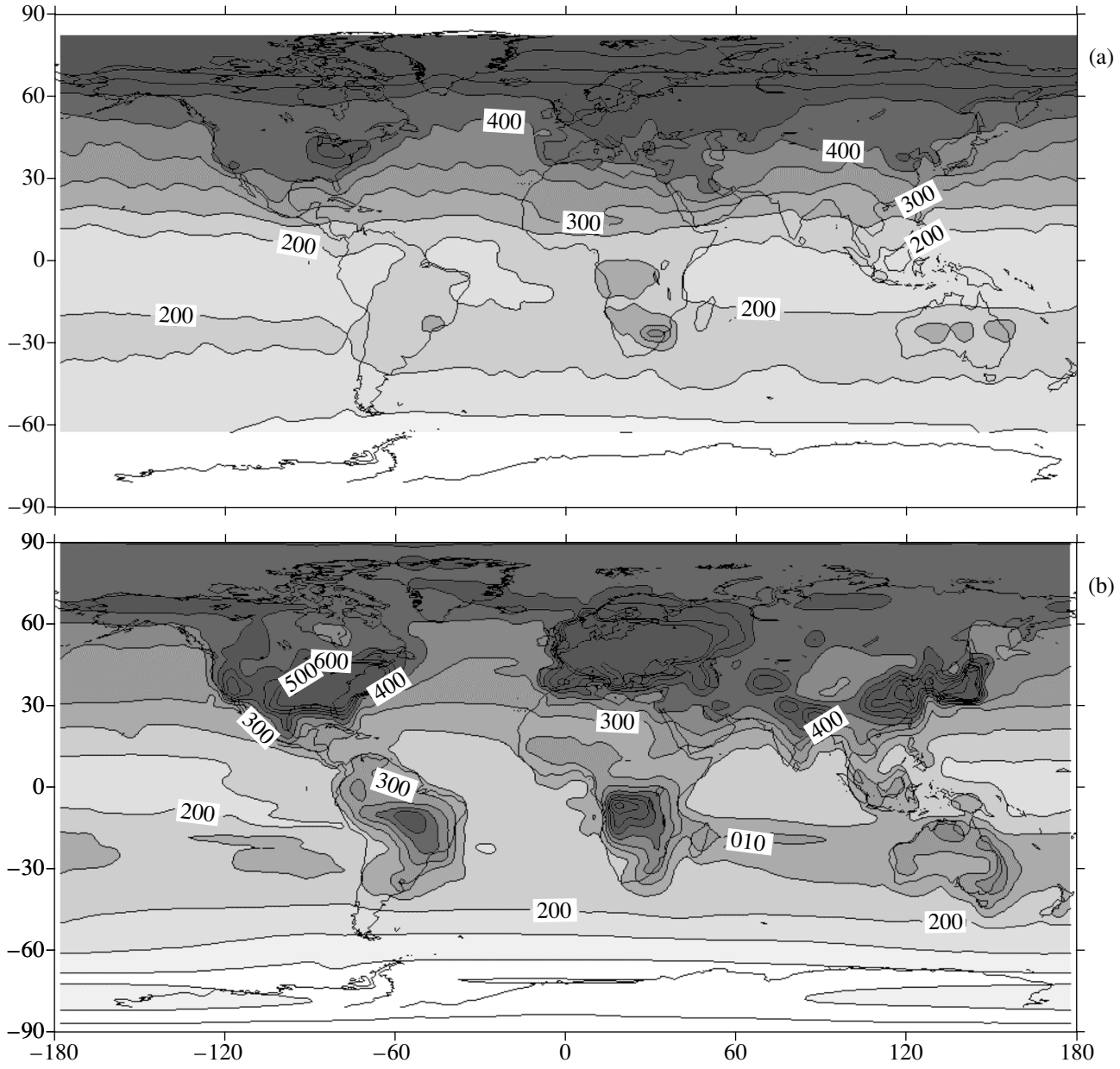

Fig. 6. Global field of the total NO_2 content (10^{13} mol/cm^2) obtained from (a) measurements and (b) simulation; July 1998.

always underestimated and this fact can serve to explain the revealed disagreement with the model.

The January and July maps of the relative discrepancies between the data of GOME measurements and MEZON simulations are presented in Figs. 3 and 4, respectively. For descriptiveness, the regions characterized by discrepancies exceeding 10% are dashed in the maps. According to comparisons with the results of independent measurements, a level of 10% corresponds to twice the mean error of GOME measurements (about 5%). It is seen that the discrepancies observed in January over most of the Earth's surface do not exceed this level; some regions located in the tropical and middle latitudes of the Northern Hemisphere and in the southeastern Indian Ocean are exceptions. The July zone of increased discrepancies between the TOC values based on the satellite measurements and model simulations covers large midlatitudinal areas of both hemispheres.The predominance of the regions for which the TOC values computed from the model exceed those obtained from the satellite measurements is characteristic of these two maps.

3.2. Global Distribution of the Total NO_2 Content

The GOME spatial distributions of the total NO_2 content have, on the whole, a zonal structure with a

maximum (about 6×10^{15} mol/cm^2) and a minimum (about 1×10^{15} mol/cm^2) falling on the high latitudes of the summer and winter hemispheres, respectively. The corresponding January and July maps obtained on the basis of the GOME measurements and MEZON simulations are presented in Figs. 5 and 6, respectively. The model reproduces the main latitudinal variation of the total NO_2 content. However, the simulated midlatitudinal NO_2 content in the Northern Hemisphere significantly exceeds the NO_2 content obtained from the observational data (especially for the winter season). The latter result is associated with the fact that, under the condition of anthropogenic pollution of the atmosphere over industrial regions of the globe, the tropospheric NO_2 content estimated by the model is overestimated in comparison with the GOME value. The maximum values of the total NO_2 content observed through the GOME measurements are equal to $(6–9) \times 10^{15}$ mol/cm^2 and are also characteristic of industrial or densely populated regions, such as western China (9.2×10^{15} mol/cm^2), northwestern and southeastern areas of the United States (8×10^{15} mol/cm^2), northern Italy (7.7×10^{15} mol/cm^2), central areas of European Russia (6.6×10^{15} mol/cm^2), and southern Africa (6.5×10^{15} mol/cm^2). Specifically, the January and July GOME measurements demonstrate some regions of increased NO_2 content, which are located in the United States, China, and central Europe (Fig. 5) and in southern Africa (Fig. 6).

It should be noted that, although the GOME data contain the total NO_2 content within the entire atmospheric column, the sensitivity of these measurements to the tropospheric NO_2 content is poorly understood. It is well known that the main portion of NO_2 is localized in the stratosphere and a significant contribution of the tropospheric NO_2 content is possible only under conditions of heavy pollution. It is obvious that clouds occurring within the field of view of satellite instruments screen the lower tropospheric layer and, therefore, special corrections based on a priori information on the atmospheric NO_2 vertical profile should be used to determine the total NO_2 content.

Table 2. Comparison of the global fields of the total NO_2 contents (T and S) retrieved from the GOME measurements performed in 1998 with those simulated on the basis of the MEZON model (mean value, rms discrepancy, and correlation coefficient; T is the atmospheric total content and S is the stratospheric total content)

Month	$\Delta_{GOME-MEZON}$, %		$\Delta_{GOME-MEZON}$, %		R	
	T	S	T	S	T	S
January	−67	14	197	23	−0.25	0.88
April	−35	2	120	16	0.48	0.83
July	−6	11	47	21	0.63	0.97
October	−29	7	113	18	0.35	0.67

The statistical estimates of the results of the comparison, namely, the mean and rms discrepancies (Δ and σ, %) between the GOME and MEZON data and the correlation coefficient R, are presented in Table 2. We compared the total and stratospheric NO_2 contents computed from the GOME measurements and MEZON simulations. On the average, the total NO_2 content com-

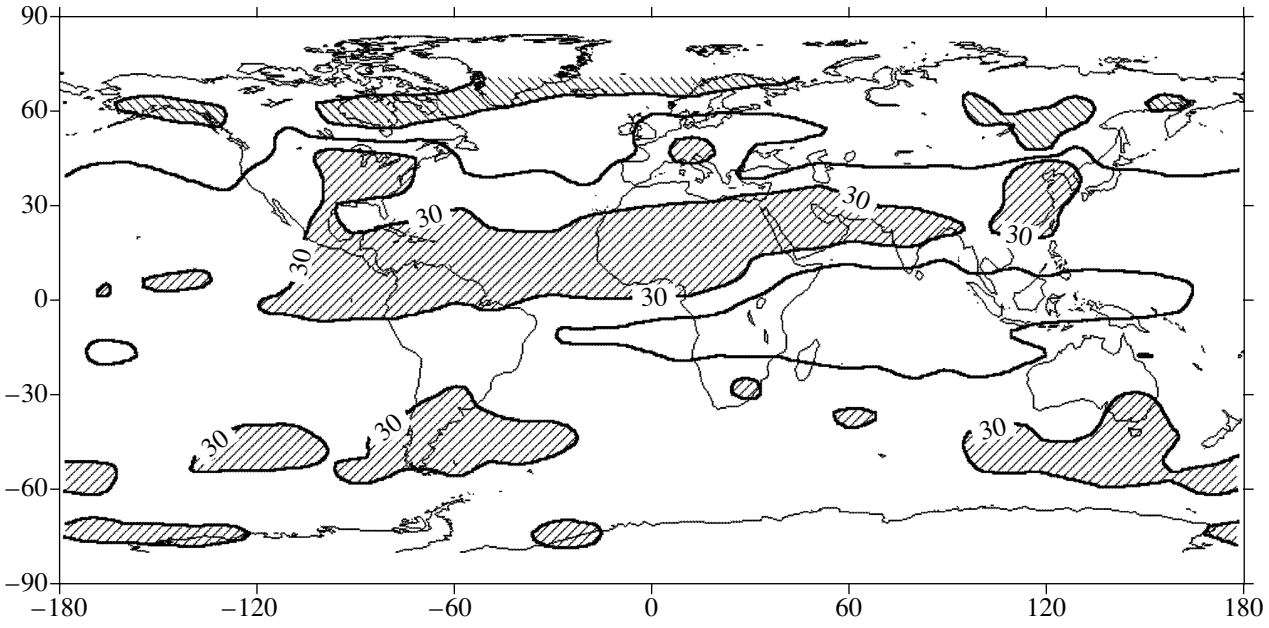

Fig. 7. Map of relative discrepancy between data on the total NO_2 content (%): (GOME–MEZON) (the stratosphere)/GOME; January 1998.

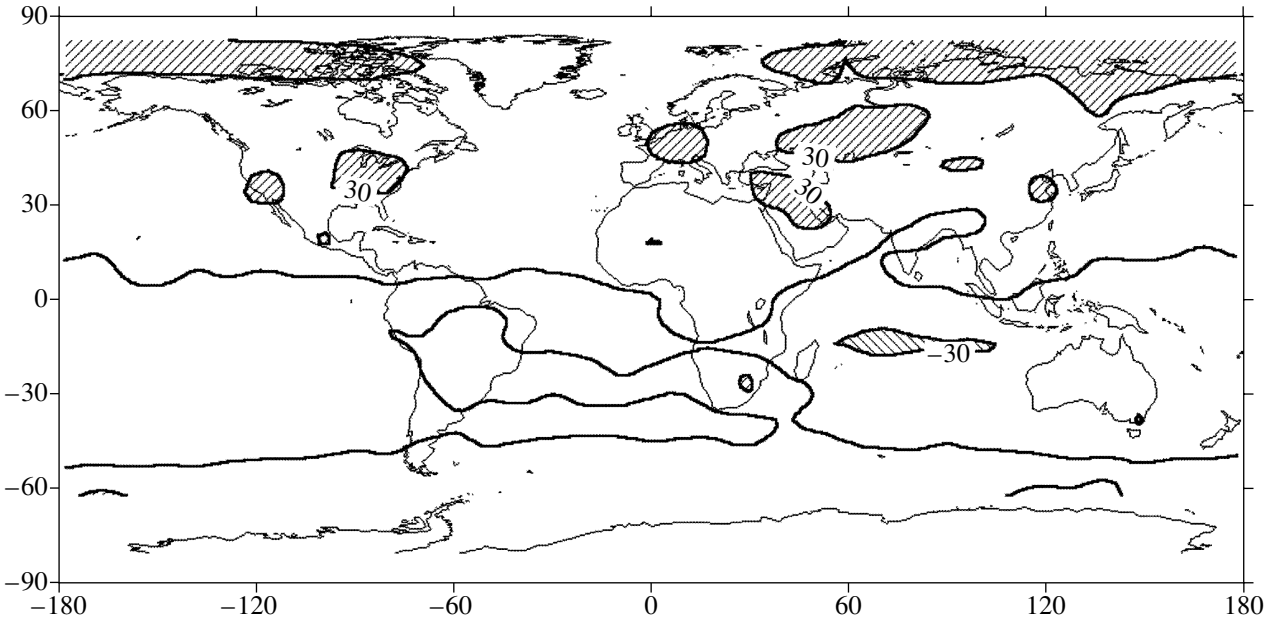

Fig. 8. Map of relative discrepancy between data on the total NO_2 content (%): (GOME–MEZON) (the stratosphere)/GOME; July 1998.

puted from the GOME measurements is significantly (by 10–70%) lower than that computed from the model, and the rms discrepancy ranges from 50 to 200%. The maximum discrepancy between the data on the total NO_2 content obtained from the GOME measurements and simulations is observed for January ($\Delta = -67\%$ and $\sigma = 197\%$). This result can be attributed to the intense accumulation of pollutants in the lower troposphere in winter. This accumulation of pollutants is completely taken into account by the model, unlike satellite measurements. The MEZON and GOME fields of the stratospheric NO_2 content correlate with each other significantly better, namely, the rms discrepancy σ ranges between 16 and 23% and, according to the comparison with independent measurements, is within twice the mean error of satellite measurements of the total NO_2 content (about 30%). The smallest discrepancies are obtained for the April data, namely, $\Delta = -2\%$, $\sigma = 16\%$, and $R = 0.83$.

The January and July maps of the relative discrepancies between the stratospheric NO_2 contents computed from the GOME measurements and MEZON simulations are presented in Figs. 7 and 8, respectively. Over the regions dashed in these maps, the discrepancies exceed 30%. It is seen that positive discrepancies are predominant for both months; i.e., the GOME values exceed the stratospheric NO_2 content values computed from the model. In January, the regions of such discrepancies have a large area and are located in the tropical and middle latitudes of both hemispheres. Some of these regions overlap such areas of heavy anthropogenic pollution of the troposphere as the United States, China, central Europe, and southern Africa (Fig. 7). A similar July map of discrepancies demonstrates that, for the high latitudes of the Northern Hemisphere, the data obtained from the GOME measurements exceed those computed from the MEZON model. On the whole, over most regions of the globe, the data based on measurements coincide within 30% with those based on simulations and the regions of positive discrepancies are located over heavily polluted regions (Fig. 8), for which the GOME data contain the contribution of tropospheric NO_2.

4. PRINCIPAL CONCLUSIONS

A comparison of the ozone and nitrogen dioxide global fields retrieved by us from satellite measurements and from simulations based on a transport–photochemical atmospheric model allows the following conclusions.

(1) The main features of the total ozone and nitrogen dioxide contents (namely, particular latitudinal distributions and local regions of increased and decreased contents) computed from the satellite GOME measurements are reproduced well through computations based on the MEZON model.

(2) The winter and spring monthly mean global fields of the total ozone content computed by the MEZON model agree with the GOME data within twice the mean error of the GOME measurements (5%), and the maximum rms discrepancy (15%) is observed for the fall season.

(3) The TOC fields computed from the model exceed those obtained from the GOME measurements by 5% (on the average), and this effect can be associ-

ated with the TOC systematic underestimation in the data of satellite measurements, which was previously noted in a number of works on GOME validation.

(4) The simulated total NO_2 contents differ significantly (by 50 to 200%) from the GOME data; maximum discrepancies are observed over the regions characterized by increased anthropogenic pollution of the troposphere and can be associated with the underestimation of the tropospheric NO_2 content in the GOME data.

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