THE SATOR INTEGRATED ECOLOGICAL PROGRAM ON STRATOSPHERIC AND TROPOSPHERIC OZONE OF THE INSTITUTE OF ATMOSPHERIC OPTICS

V.V. Zuev

Institute of Atmospheric Optics,
Siberian Branch of the Russian Academy of Sciences, Tomsk
Received April 5, 1992

The photochemical reactions and their reactants affecting the ozone content in the stratosphere and troposphere are studied. The set of the parameters and components of the troposphere and stratosphere is identified which must be determined for integrated study of the dynamics of the Earth's ozone layer. The structure of the SATOR integrated program on stratospheric and tropospheric ozone is described.

The increasing amount of emissions of the harmful substances in the atmosphere, hydrosphere, and on the planetary surface caused by a forced industrialization of our society leads to the noticeable deterioration of the ecological state of the environment. Transboundary transports of the pollutants, especially in the atmosphere, exhibit the planetary scale; therefore, the problems of ecological monitoring and protection of the environment have become the primary international problems. Attention of the whole mankind is attracted to such global atmospheric phenomena as the greenhouse effect and ozone holes. Intensification of the greenhouse effect is associated with increase of the concentrations of such gases as CO_2, CH_4, N_2O, O_3, freons, and others which absorb the upwelling thermal radiation emitted by the heated Earth's surface. The greenhouse effect provokes steady warming–up of the surface layer of the atmosphere and underlying surface which may lead to global changes of the planetary climate, melting of the polar ices, and rise of the global ocean level.

The ozone holes are characterized by a sharp depletion of the total ozone content (TOC) in the atmosphere because of destruction of the ozone layer in the stratosphere. Depletion of the TOC promotes the penetration up to the ground of the short–wave part of the UV solar radiation in the wavelength range 290...320 km, being destructive for all biological forms of planetary life. For the last decade the ozone holes are regularly recorded over the Antarctica in spring. At this period the TOC falls off sometimes by more than twofold in comparison with its normal quantity.

The long–term observations of the vertical distribution of ozone in the Nothern Hemisphere performed at the Hohenpeissenberg observatory in Germany[1] showed (see Fig. 1) the steady decrease of the ozone concentration in the maximum of the stratospheric ozone layer and, on the contrary, the increase in the tropospheric ozone concentration. On the whole, these trends characterize the decrease of the TOC over the Europe because of predominating contribution of the ozone content in the maximum of the stratospheric ozone layer to the TOC. At present these trends in the TOC at high and middle latitudes of the Nothern Hemisphere are getting more pronounced due to the depression of the stratospheric ozone layer after eruption of the Pinatubo volcano in Philippine occurred from July 13 to July 15, 1991. The most substantial decrease of the ozone content was observed at the end of January of 1992 over the Nothern Europe (Fig. 2). The deviation of the TOC from the long–term norms reached 45%. As a matter of fact, Fig. 2 shows the formation of the ozone hole over the Europe. On the whole, the zone of the substantial depression of the stratospheric ozone layer covers the Nothern Atlantics, Europe, and Western Siberia.

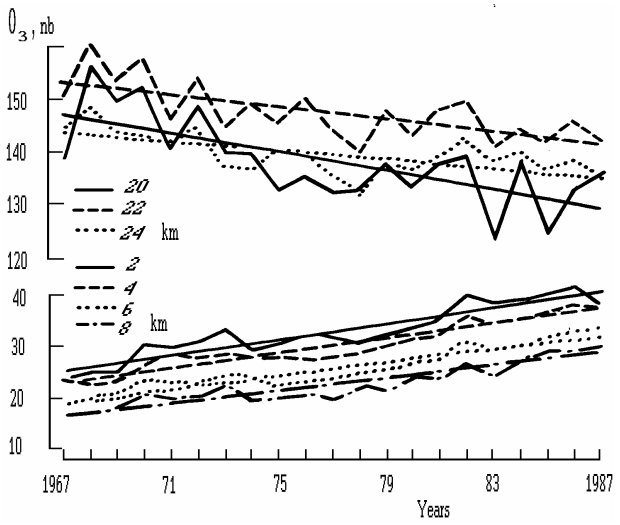


FIG. 1. Trends in variation of the stratospheric and tropospheric ozone concentration according to the data of the 20–year observations of the vertical ozone distribution performed at the Hohenpeissenberg observatory.[1]

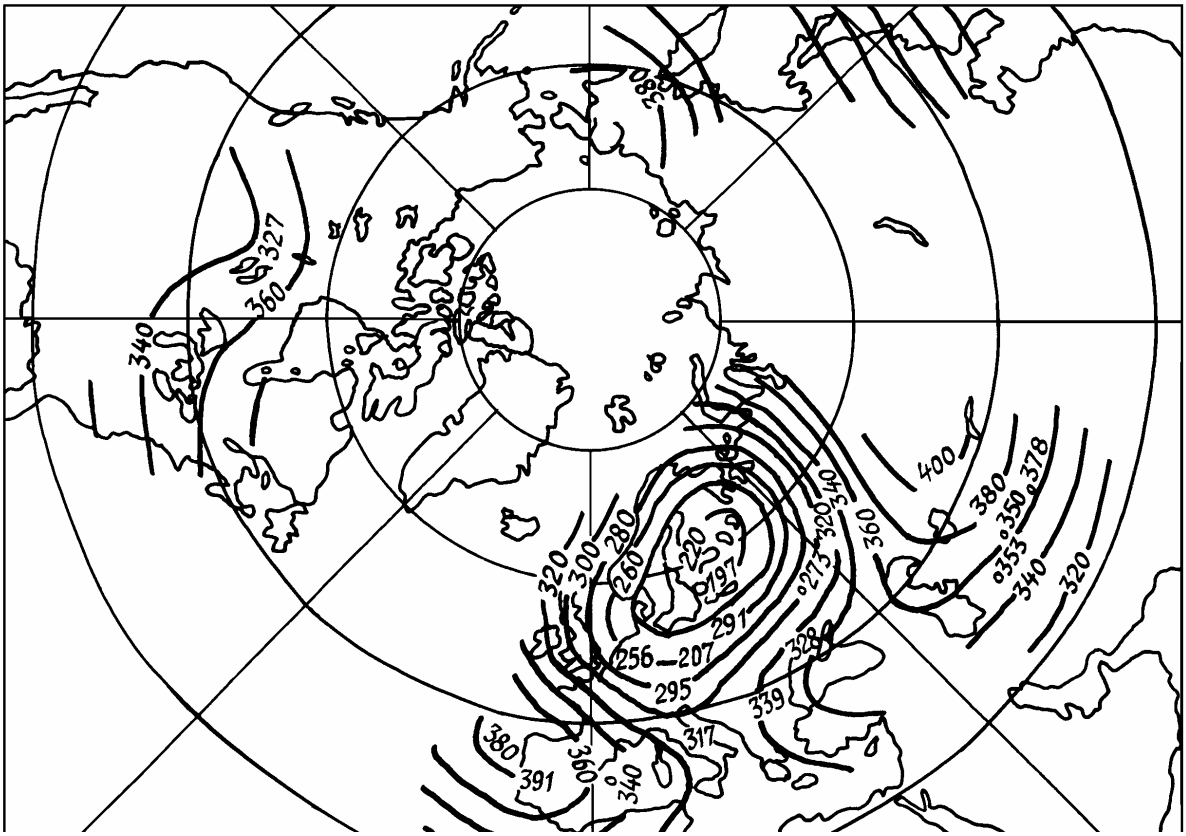
The mechanism of forming the ozone holes is ambiguous. At least, it differs in the Southern and Nothern Hemispheres due to specific features of the circulation of the air masses. However, undoubtedly a significant contribution to these processes comes from the photochemical reactions especially the reaction of the catalytic decomposition of ozone by the Cl atoms and radicals NO or CH. The chlorine atoms produced in the stratosphere as a result of the photodissociation of freon molecules upon exposure to the UV solar radiation actively decompose ozone according to the scheme[2]

$$Cl + O_3 \rightarrow ClO + O_2 ; \quad (1)$$

$$Cl + O \rightarrow ClO + O_2 , \quad (2)$$

0235-6880/92/06 354-03 $02.00

Total ozone concentration (Dobson units) on January 28, 1992



Total ozone concentration (Dobson units) on January 29, 1992

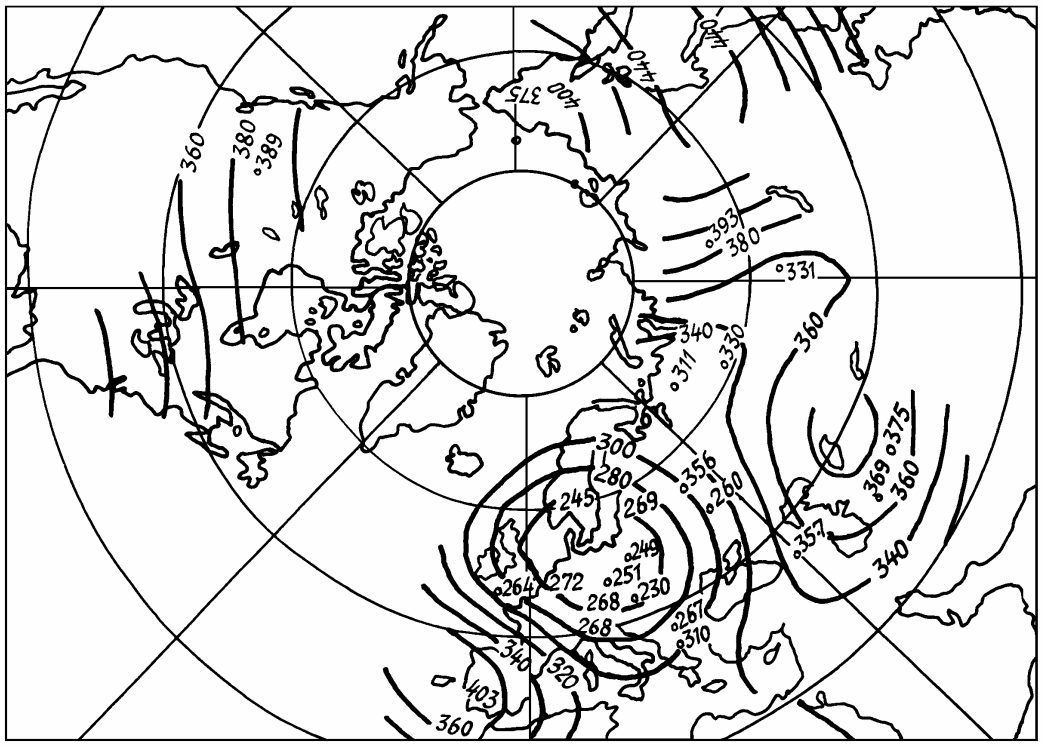


FIG. 2. The ozone hole over the Nothern Europe caused by the depression of the ozone layer after the eruption of the Pinatubo volcano.

which shows that each atom of chlorine is capable of decomposing several thousands of the ozone molecules until it recombines into the stable molecule–reservoir, for example, $ClONO_2$. On the whole, to determine the chlorine balance in the atmosphere, simultaneous monitoring of three substances: HCl, ClO, and $ClONO_2$ is needed.

Analogously, NO and NO_2 take part in the catalytic reaction of decomposition:

$$NO + O_3 \rightarrow NO_2 + O_2 ; \quad (3)$$

$$NO_2 + O \rightarrow NO + O_2 , \quad (4)$$

in addition, reaction (3) predominates in the processes controlled by stratospheric ozone; therefore, obtaining the exact data on the ratio NO_2/NO has primary importance for description of the ozone cycle. Since after reaction (3) photolysis of NO_2 molecule can take place and by it an odd oxygen is formed

$$NO_2 + h\nu \rightarrow NO + O , \quad (5)$$

the rate of the catalytic decomposition of ozone according to scheme (3)–(4) depends on the rates of reactions (4) and (5). NO is produced in the stratosphere mainly due to decomposition of nitrous oxide N_2O reacting with the excited oxygen atom O ('D). To evaluate the nitrogen cycle in the atmosphere, simultaneous information about all the active nitrogen compounds: NO, NO_2, NO_3, N_2O_5, HNO_3, HO_2NO_2, and $ClONO_2$ which are referred to as odd nitrogen NO_x, is needed.

Catalytic decomposition of ozone in the atmosphere, in which hydroxyl radical OH takes part, proceeds according to the scheme described by Eqs. (1) and (2) or (3) and (4). However, in the region of the tropopause and below in the troposphere the reaction

$$OH + O_3 \rightarrow HO_2 + O_2 ; \quad (6)$$

$$HO_2 + O_3 \rightarrow OH + 2O_2 , \quad (7)$$

in which ozone takes part twice, is most effective.[2] The content of OH in the atmosphere is mainly determined by photochemical reactions in which the key role is played by methane (CH_4). To estimate the content of hydroxyl OH in the atmosphere, we may use the ratio HCl/HF since, in contrast to HCl, the molecule HF does not react effectively with OH while their sources are approximately the same.

Along with the steady increase of the freon content in the atmosphere (mainly freon–11 and freon–12 used in the refrigerators and aerosols) leading to an increase of the concentration of the chlorine atoms in the stratosphere, the halogens (1301 and 1211) used for extinguishing, are also accumulated in the atmosphere. They photodissociate producing the bromine atoms (Br). A bromine atom not only takes part in the catalytic decomposition of ozone according to the scheme analogous to reactions (1) and (2), but also affects the chlorine cycle by means of reaction with ClO (see Ref. 2)

$$BrO + ClO \rightarrow Br + OClO \rightarrow Br + Cl + O_2 , \quad (8)$$

which intensifies the effect of both halogens (Br and Cl) on the decomposition of ozone.

The above–considered processes associated with the ozone cycle in the stratosphere indicate that for its description we must simultaneously carry out the integrated monitoring of a large number of the parameters and components of the stratosphere. These are wind, temperature, pressure, and such gaseous components as O_3, NO, NO_2, N_2O, HNO_3, N_2O_5, H_2O, CH_4, CO, CF_2Cl_2 (freon–12), $CFCl_3$ (freon–11), HCl, ClO, $ClONO_2$, HF, HBr, and BrO. In addition, the stratospheric aerosol through the surface of which the ozone sink takes place, must be simultaneously monitored. Under conditions of large aerosol perturbation in the stratosphere after volcanic eruptions this process can make a significant contribution to the depression of the ozone layer, as was the case after eruption of the Pinatubo volcano.

The increase of the tropospheric ozone concentration over the Europe (see Fig. 1) is associated with the photochemical transformation of gases of the industrial origin. The danger of the outlined tendency, in spite of the small values of the tropospheric ozone concentrations, is its high toxicity and its contribution to the greenhouse effect. The ozone cycle in the troposphere is determined by a large number of chemical reactions in which various gases take part. To describe the ozone cycle, simultaneous information about ozone, NO, NO_2, NO_x, CH_4, CO, CO_2, $C_nH_{2n+2}(n \geq 2)$, freons, J_{NO_2}, $J_{O('D)}$, aldehydes, ketones, PAN (peroxydeacetyl nitrites), H_2O_2, and meteorological parameters of the troposphere is needed.[3] Representation of these data, because of the fast dynamics of the air flows in the lower troposphere, greatly depends on the simultaneous monitoring of the nearest sources of pollutants in the atmosphere and of the wind–driven transport of these pollutants in the boundary layer of the atmosphere. The powerful natural source of the ozone emissions in the troposphere can be the tropopause breakthrough and sedimentation of the ozonized stratospheric air masses. The maximum recorded ozone concentration in the troposphere are associated exactly with this effect (see, for example, Fig. 3), which, naturally, occurs occasionally.

The aerosol sink of ozone in the lower troposphere, especially within the roughness layer, as a rule, is much less then the sink on the ground, buildings, and forests stands. However, under conditions of intensive emissions of industrial or natural aerosol (for example, of dust storms) their effect on the ozone sink can be significant.

Generalizing the total set of the parameters and components of the atmosphere, directly or indirectly influencing the ozone balance in the stratosphere and troposphere, it is easy to see that in many respects it determines the radiation and heat balances in the atmosphere including the greenhouse effect. Thus the integrated long–term monitoring of the tropospheric and stratospheric ozone and of the components of the ozone cycle on the basis of this set of the parameters has also the climatological character.

In 1991 the SATOR long–term integrated program on stratospheric and tropospheric ozone was started at the Institute of Atmospheric Optics. The main idea of this program is to perform the long–term integrated experiment in the atmosphere using a large number of various devices located at one place, to obtain, and to analyze comprehensively the long series of synchronous atmospheric observations. The structure of the SATOR program is shown in Fig. 4.

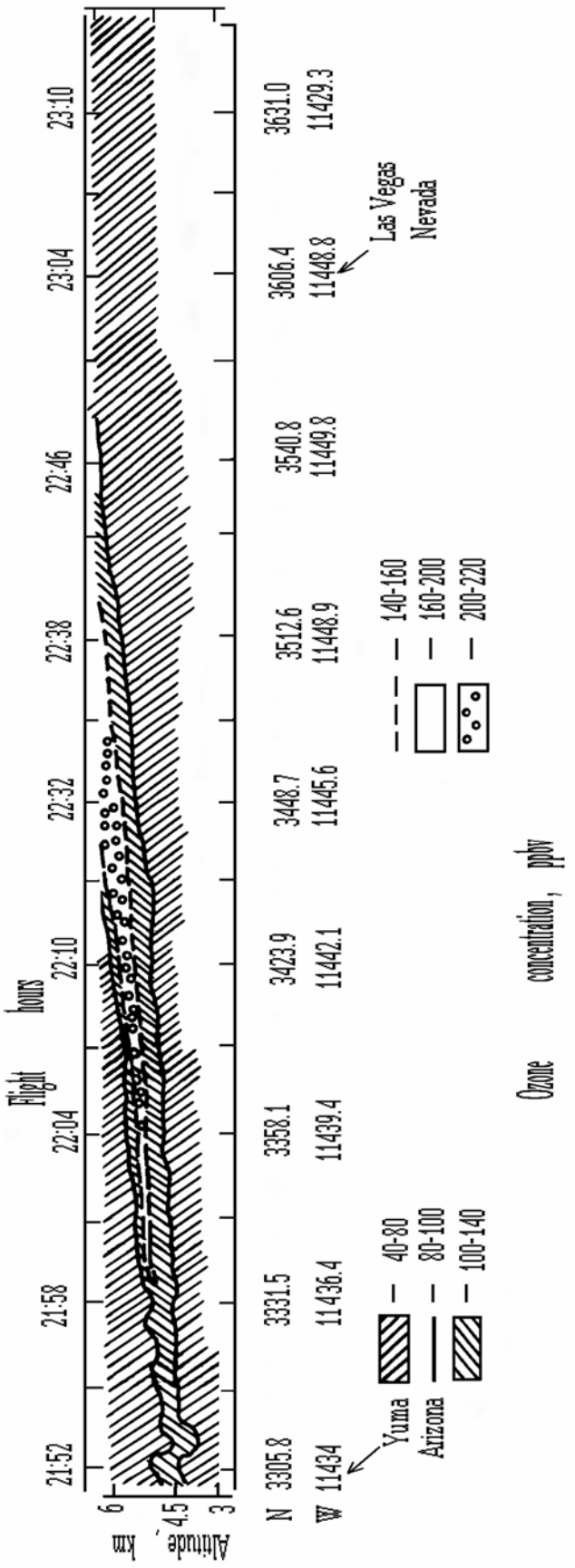


FIG. 3. Recording of the stratospheric ozone breakthrough into the troposphere.[1]

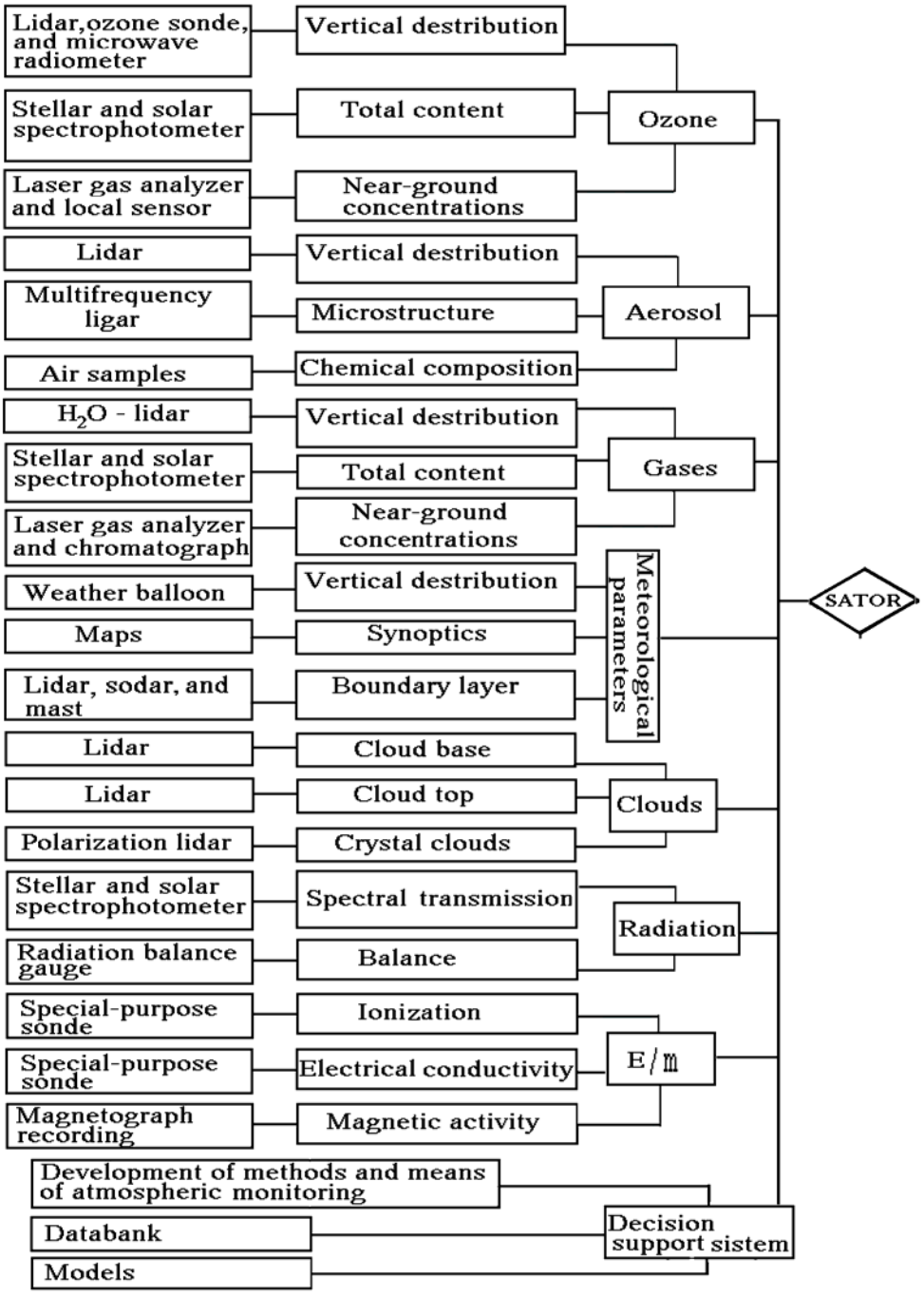


FIG. 4.

The considerable accent in this program, with regard to the distinguishing features of the Institute of Atmospheric Optics is made on the use of the methods of laser and optical monitoring of the atmosphere by means of lidars and spectrophotometers. In addition, we use the acoustic sounding and contact methods including the direct intake of the air samples with subsequent analysis of their physical and chemical compositions. The main elements of the program are the first four blocks: "Ozone", "Gases", "Aerosol", and "Meteorological parameters". In the block "Ozone" the UV lidar gives an information about the ozone profile in the 0–40 km altitude range. At present we have mastered the 10–30 km altitude range alone. The measurements were carried out only at night because of the limitations due to background and, naturally, under cloudless conditions. The continuous observations are possible in the 0–15 km altitude range. The ozonesonde and microwave radiometer operate continuously and are practically all–weather devices. The radiometer gives an information about the ozone content in the 25–60 km altitude range. In the block "Gases" we will reconstruct their

total content in the vertical atmospheric column using the data of the stellar and solar spectrophotometer. Aerosol data including their microstructure are obtained at the altitudes up to 50 km. Blocks "Clouds" and "Radiation" in which the information is obtained practically with the help of the same devices as in the main blocks are attendant. But with these blocks the program acquires the climatological aspect. It should be underlined that the most complete and interesting data can be obtained here with the use of the unique possibility of the aircraft laboratory of the Institute of Atmospheric Optics equipped with lidar, spectrophotometer, radiometer, and contact sensors. As a matter of fact, the use of the aircraft laboratory will give us an opportunity to perform the CRE (Complete Radiation Experiment). Unfortunately, the operation of this aircraft laboratory is too expensive.

The E/M block (of the electric and magnetic activity of the atmosphere) is associated with an account of the effect of the solar–terrestrial relationships on the atmospheric processes and ambiguity of the interaction of ozone with charged or neutral particles.

And, finally, in the block "Decision support system" we develop the new systems for monitoring of the atmosphere with time of their development not longer than one year and create the informational databank accumulating the results of the integrated experiment for the integrated cross–analyses being investigated.

The SATOR program has to be completely operated by the end of 1992.

REFERENCES

1. K. Wege, H. Claude, and R. Hartmannsgruber, in: *Ozone in the Atmosphere*, R.D. Bojkov and P.A. Fabian, eds. ISBN 0–937 194–15–8. Deepak Publishing (1989), pp. 109–112.
2. *The Atmosphere. Handbook* (Gidrometeoizdat, Leningrad, 1991), 510 pp.
3. *Eurotrack Annual Report*, Part 9, TOR (1989).
4. E.V. Browell, E.F. Danielsen, S. Ismail, G.L. Gregory, and S.M. Beck, J. Geophys. Res., No. 92, 2112 (1987).