A-1 Research on the factors to influence the future ozone layer

Contact person Takahsi Imamura
Head, Ozone Layer Modeling Research Team
National Institute for Environmental Studies
Onogawa 16-2, Tsukuba, Ibaraki 305-8506, Japan
Tel: +81-298-50-2406 Fax: +81-298-50-2579
E-Mail: Imamura@nies.go.jp

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1. Introduction

The efforts under the Montreal Protocol and its various Amendments recently led to declining the total concentration of organic halogen compounds in the stratosphere. However, the recovery of ozone layer has not yet been detected. The future state of ozone layer must be controlled by the stratospheric concentrations of man-made halogen compounds, but would be affected by changing the atmospheric abundances of greenhouse gases, natural trace gases, water vapor, and sulfate particles as well as by climate change. To elucidate the influence of these factors on future change of stratospheric ozone, the following investigations are required: (1) the detection of any trend or significant variability in the chemical and/or physical parameters in the stratosphere, (2) development of numerical models including stratospheric processes and numerical experiments of the stratospheric response against the changes of possible controlling factors, e.g., the increase of CO_2, and (3) the determination of physical and chemical parameters to evaluate the proposed hypotheses.

This project consisted of five groups and the following studies were carried out during above fiscal period.

1. Variability of trace gases and polar vortices and its impact on ozone destruction

Long and short term variability in polar vorticity was calculated and visualized for both the Arctic and Antarctica. Ozone depletion and diabatic descent in the Arctic polar vortex was studied with a diabatic trajectory model. Ozonesonde measurements were carried out over east Siberia to evaluate the ozone loss in the vortex edge region.

2. Effects of reduction of stratospheric temperature on composition of and heterogeneous reactions on polar stratospheric clouds

The transmission and reflection FTIR spectra of model PSC films were observed. A negative-ionization mass spectrometer was developed to obtained kinetic information of halogen oxide radical reactions.

3. Modeling on the response of ozone to changing atmospheric emissions of source gases and greenhouse gases and to climate change

A photochemical-radiative 1-dimensional coupled model, a 3-dimensional chemical transport model, and an atmospheric general circulation model with coupled stratospheric chemistry were developed to understand the mechanisms of stratospheric processes and to investigate the future changes of stratospheric ozone. Kinetic data for proposed heterogeneous reactions on sulfuric-acid aerosols were measured.

4. Analytical research of the ozone variation factor at the mid-latitude based on the ozonesonde observations

The statistical analysis of ozonesonde data over Tsukuba was carried out to estimate how the height of the tropopause, QBO, and solar activity influence on ozone in the mid-

mid-latitude.

5. Analysis of satellite and balloon data with a photochemical model

A trajectory chemical model was developed to estimate the ozone loss rates in the Arctic vortex. The model was applied to analyze the ILAS data and ozonesonde data over east Siberia and Hokkaido.

The following results are highlighted.

2.1. Long and short term variability in Arctic polar vortex indices

If the polar vortex is stable for long period, polar air is essentially isolated from midlatitude air masses, hence ozone loss in the vortex would become cumulative. The polar vortex in the Northern Hemisphere has been believed to not to be well formed and as stable as that in the Antarctica. This trends to cut short the processes that destroy ozone in the Arctic. However, satellite observations suggest that total ozone values have been decreasing in the Arctic, especially since the late 1980s. Therefore, we should answer a question; does the Arctic polar vortex become strong and/or stable during this one or two decades?

Ertel's potential vorticity (PV) distributions on isentropic surfaces are useful for viewing the dynamical processes related to the Arctic polar vortex because PV behaves as a quasi-conserved tracer. PV can be calculated using temperature and wind fields from three-dimensional global analysis data from the ECMWF, NCEP/NCAR, etc. To eliminate the dependence of PV on potential temperature and equivalent latitude, ϕ_e, the normalized gradient of PV is defined as NGPV=(dPV/d ϕ_e)/PV.

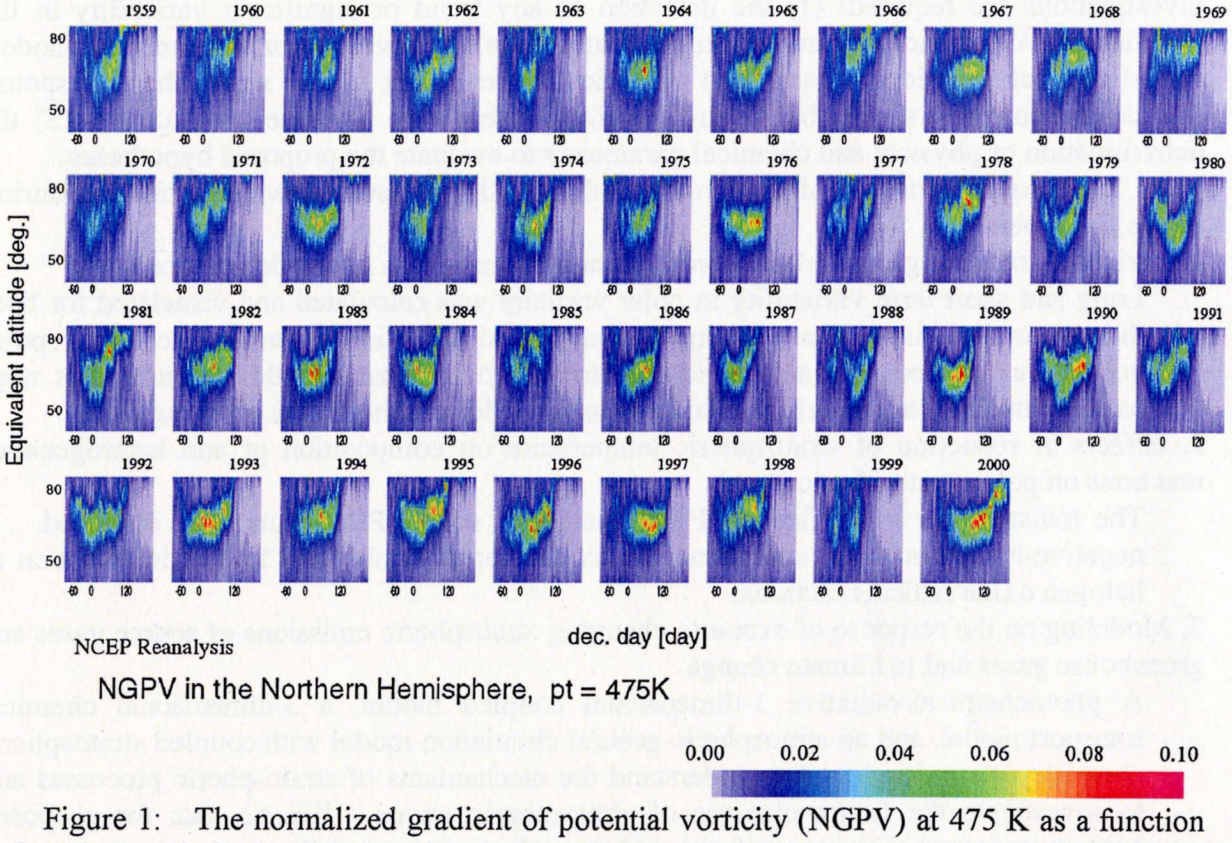

Figure 1: The normalized gradient of potential vorticity (NGPV) at 475 K as a function of equivalent latitude and time for the 42 winters from 1959 to 2000 from 45°N to 85°N calculated using the NCAR/NCEP re-analysis data. (Ref. 1)

NGPV was plotted as a function of equivalent latitude and time for the 42 winters from 1959 to 2000 in the Northern Hemisphere (see Figure 1). The plots made it easy to

extract any positive or negative trends in the strength, duration, size and stability of the polar vortex. The following characteristics were found through the quantitative analysis of the vortex indices;

(1) There were positive trends in the strength, duration, radius and stability of the vortex.
(2) Short-term variability in the indices was correlated with the QBO.
(3) A coupling of the 11-year solar cycle and the QBO generated long-term variability. Positive correlations were found between all four indices (strong correlation in the case of vortex strength) and the QBO during solar inactive phases. During solar active phases, the correlations were weak or weakly negative.

2.2. Lagrangian estimation of ozone depletion in the Arctic polar vortex 1995/1996

Ozone depletion and diabatic descent in the Arctic polar vortex in winter 1995/1996 was studied with a diabatic trajectory chemistry model developed by NIES. To study the chemical and dynamical evolution of the species in the polar vortex, 400 diabatic trajectories were calculated in the vortex core and vortex edge region, by using three-dimensional (3-D) winds data provided by the European Centreer for Medium-Range Weather Forecasts (ECMWF). Ozonesonde data from two stations (Ny-Alesund in the vortex core and Yakutsk in the vortex edge) were compared with the model results. Ny-Alesund was mostly located in the vortex core, while Yakutsk were mainly in the vortex edge region. Good agreements were obtained between the model results and observations for both the vortex core and vortex edge region. These results suggest that the strong ozone depletion occurred not only in the vortex core, but also in the edge region of the vortex, and that air masses from the mid-latitudes did not considerably affect the degree of ozone depletion in this winter/spring period. The model sensitivity to different descending rates and to the presence of large NAT particles was examined.

2.3. Impact of ozone depletion in the Arctic polar vortex on ozone in the northern mid-latitude

Ozone depletion in the Arctic have been decreasing, especially, since the late 1980s. If the ozone-depleted air in the polar region significantly mixed into the air in the mid-latitude, the total ozone in the mid-latitude would partly be perturbed by the mixing. To detect the airmass with the polar character over the mid-latitude, vertical profiles of ozone have been measured at Moshiri (44 °N, 142°E) in Hokkaido, Japan since 1996. Episodic low ozone events were found on April 14 and 23, 1996. According to the meteorological analysis, the station was found to be inside the Arctic polar vortex at that time. Significant chemical ozone loss was found in the Arctic vortex in this year by the MATCH technique. The ozone profile obtained at Moshiri on April 23, 1996 was compared with the profile measured at Sapporo on April 13, 1972 when Moshiri and Sapporo (43 °N, 121 °E) were inside the Arctic polar vortex. The dynamical condition of the Arctic polar vortex in 1972 was similar to 1996 except that the vortex was a little weaker in 1972 and the temperature inside the vortex was higher in 1972. The ozone profiles at Sapporo in 1972 were similar to those at Moshiri in 1996. At around 70 hPa, the ozone mixing ratio was 55% smaller in 1996 at Moshiri compared to 1972 at Sapporo case.

The Improved Limb Atmospheric Spectrometer (ILAS) observed low-concentration N2O regions in high latitudes of the lower stratosphere after the Arctic vortex breakdown in May 1997. A new chemical transport model developed (referred to as the CCSR/NIES nudging CTM) was used to simulate the observed low-N_2O air masses, because N_2O is a good tracer of the polar air: i.e., lower mixing ratio of N_2O in the polar vortex than in the mid-latitude. The model could simulate the observed time-longitude corss-sections of N_2O mixing ratio. The simulation showed that one of the air masses had a horizontal scale of 1,000-1,500 km, and remained at high latitudes for 1.5-2.5 months after the polar vortex

breakdown. The simulation also showed that the contrast between the N_2O concentration of the low-N_2O air mass and that of the ambient air was diminished when the air mas was stretched, which had a considerable influence on the duration of the air mass.

2.4. Impact of the increase of CO_2 on stratospheric ozone

CO2 is chemically inactive in the atmosphere and its concentration is also increasing in the stratosphere. The increase of CO_2 is expected to produce a cooling of the stratosphere. Chemical-radiative one-dimensional model calculations showed that the cooling was reduced by as much as 30% at altitudes above 40 km due to the increase of ozone. The calculations also found that the change in the solar ultraviolet radiation in the stratosphere due to the ozone increase had considerable effects on the concentrations of chemical species, such as NO_y, ClO_y, etc.,

Because of feedbacks between chemistry, radiation and dynamics in the stratosphere, the impact of a CO_2 increase on the stratospheric ozone would not be simple and, hence, analysis with a fully coupled model is desired. We developed and improved an atmospheric general circulation model with fully interactive stratospheric chemistry (referred simply as to CCSR/NIES AGCM) for the numerical integration of ozone change against the increase of CO_2. The amount of the minimum column ozone in the ozone hole and the size of the ozone hole calculated by the model were in good agreement with the TOMS data.

2.5. Simulation of stratospheric sulfate aerosols.

The photochemistry for SO_x species was included in the CCSR/NIES AGCM to investigate the impact of a volcanic eruption on stratospheric aerosols and ozone. The model was used to simulate the distribution of background stratospheric sulfate aerosols. The numerical results demonstrated that the model well simulated the distribution of the surface area density of sulfate aerosols in the lower stratosphere (see Figure. 2). The model implied that the SO_2 emission at the surface can affect the stratospheric sulfur burden.

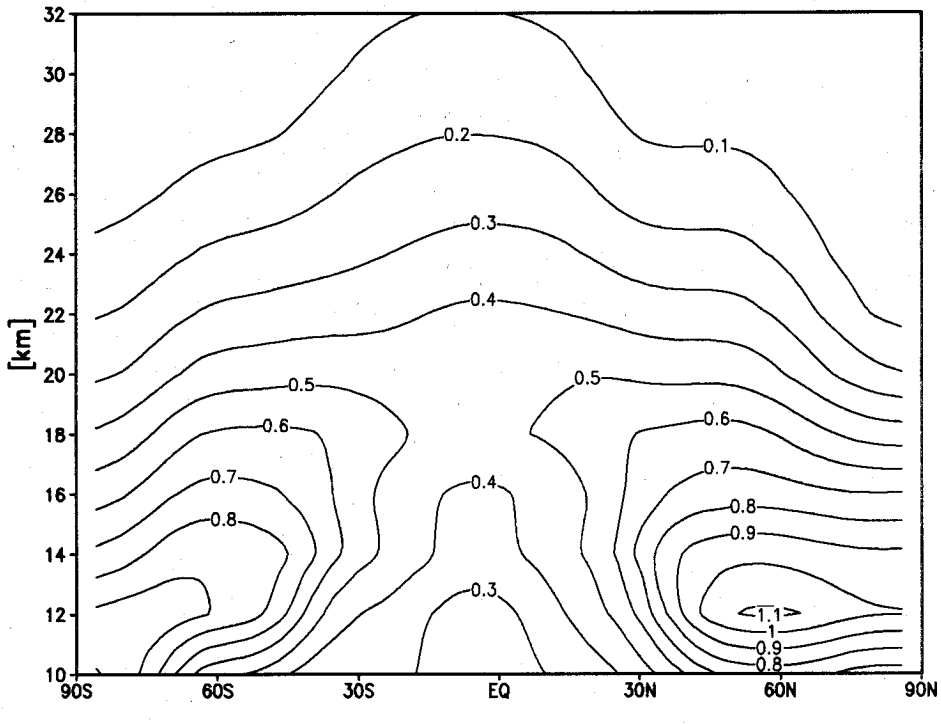

Figure 2: Calculated latitude-height cross section of the zonally averaged monthly mean aerosol area density in March. The contour interval is 0.1 $\mu m^2/cm^3$. (Ref. 2)

Simulation of stratospheric sulfate aerosols under a volcanic condition was also carried out by using the CCSR/NIES AGCM. The numerical results showed that the temperature in the lower stratosphere increased after the eruption. The model with the heterogeneous reactions on the aerosols could reproduce the observed warm temperature anomaly for about one year after the eruption. The model results indicated that the negative anomaly of ozone observed were related to heterogeneous reactions on the sulfate aerosols.

2.6. Heterogeneous reactions on sulfate aerosols

HC(O)OH in the stratosphere is thought to be a product of a newly proposed heterogeneous reaction of formaldehyde in sulfuric-acid aerosols: $H_2CO+HNO_3 \rightarrow HC(O)OH + HONO$. A rotatable wetted-wall flow reactor was used to observe the uptake behavior of HC(O)OH into sulfuric-acid solutions. It was found that the uptake of HC(O)OH was reversible and that the reactive loss of HC(O)OH in the solution, $HC(O)OH \rightarrow CO + H_2O$ was negligibly slow. The Henry's solubility of HC(O)OH in the solution was obtained. The results indicated that HC(O)OH would be a good tracer of the heterogeneous reaction of H_2CO with HNO_3 in sulfuric-acid aerosols.

3. References:

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