A-1 Research on Explanation of Long-Term Trend and Prediction of Future Change of Ozone Layer

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1. Introduction

Owing to the regulation of ozone-depleting substances, such as chlorofluorocarbons and bromofluorocarbons, the stratospheric abundances of chlorine and bromine compounds are believed to have started to decrease. It is expected that the stratospheric abundances of halogens (chlorine and bromine) will return to the 1980 amounts during the middle of this century. However, the atmospheric abundance of several other species influenced by human activities, such as CO_2, methane, nitrous oxide, and the meteorological conditions, has changed from that in the 1970s.

Water vapor measurements in the stratosphere are limited and the trend of stratospheric water vapor has not yet been definite. However recently, it is reported that the amount of water vapor in the lower stratosphere may increase between 1980 and 2000 with the increase rate of ~1%/yr.[1)] If the reported increase rate is correct, such a high increase rate can not be explained by the effect of CH_4 oxidation and, then, the increase of stratospheric water vapor should be due to the change of dynamical processes. Water vapor is a source gas of the aerosols and HO_x radicals in the stratosphere.

There is not enough evidence to support the idea that the ozone layer would recover to a normal state if man-made chlorine- and bromine-containing compounds decrease because the change in atmospheric compositions affects the transport processes and/or the production and destruction rates of ozone in the stratosphere.

2. Research Objective

The aims of this research projects are: (i) development and improvement of numerical models, chemical climate model (CCM) and chemical transport model (CTM), including stratospheric processes, (ii) to examine the response of stratospheric ozone to the increase of CO_2 with the CCM, (iii) detection of the trend of stratospheric water vapor in the tropical tropopause layer region where the amount of water vapor transported from the troposphere into stratosphere is controlled, (iv) measurements of the chemical reaction data related to the HOx cycle, and (v) understanding of the

ozone trend in the mid-latitude.

3. Results and discussion:

This research project consists of three main groups and an Eco-Frontier subgroup. The following studies were carried out during the period of this project.

(i) Prediction of changes in ozone with increasing CO_2:

A chemical climate model, an atmospheric general circulation model with fully interactive stratospheric chemistry, with T21 horizontal resolution (T21-version CCM) was developed. The model was used for the Sixty-five year numerical integration of ozone change against the changing of the concentration of CO_2 and organic chlorine compounds. It was found that ozone hole event is not sensitive to the CO_2 concentration but appears to be controlled by simulated chlorine loading. However, the model could not well reproduce the period of severest ozone depletion in the ozone hole. That is, predicted period of the severest ozone depletion was delayed by about one month more than that in the TOMS observation.

After a future ozone prediction calculation using the CCSR/NIES CCM with T21 horizontal resolution, a CCM with T42 horizontal resolution is developed. The T42-version model includes the gravity wave parameterization proposed by Hines and bromine chemistry. The effect of the Earth's sphericity has also been taken into account in the new-version model. The heterogeneous reaction scheme, the chemical kinetics and photochemical data were updated. In the new version of the CCM, the cold bias in the winter polar lower stratosphere is improved with a zonal mean minimum temperature of 180 K.

(ii) Stratospheric water vapor change - Mechanisms, trends, and their impact on stratospheric ozone:

Accumulation of the observational evidences on the water distribution is a key to improve our understanding on the long-term trend of the water vapor in the Tropical Tropopause Layer and the lower stratosphere. Campaign observations were conducted in the central to the western tropical Pacific under the framework of SOWER during the period of this project. We could see that the water vapor distributions show characteristic features between each station reflecting the progress of dehydration for the advecting air parcels.

A low-cost, chilled-mirror hygrometer for radiosonde applications, named "Snow White" hygrometer, was compared with H-Humicap sensor from Vaisara and NOAA/CMDL frostpoint hygrometer. These intercomparisons confirm the validity of the Snow White measurements at least up to the tropical upper troposphere and above 3%-6% RH. The comparison between the Snow White and A-Humicap sensors shows the known A-Humicap dry bias error at low temperatures and second dry bias error in the wet low troposphere.

The role of large-scale wave-driven dehydration as a mechanism to control the water vapor in the tropical tropopause layer region was investigated using the CCSR/NIES AGCM. In the model calculation, eastward moving large-scale equatorial gravity waves are found to be dominant to modulate the distribution of the minor constituents around the equatorial tropopause. It was suggested by a case study over the Indian Ocean in the northern summer that the equatorial Kelvin

wave plays a role in the dryness in the equatorial tropopause region.

The formation of electronically excited atomic oxygen, $O(^1D)$ and $O(^1S)$, from the photolysis of O_3 was measured by means of a tunable laser photolysis combined with vacuum UV laser-induced fluorescence detection of the atoms. New photochemical data on ozone photolysis were obtained.

(iii) Analysis of factors that influence ozone in the mid-latitude

The variation at mid-latitude ozone is caused by various factors and the variation processes are complicated. They are caused not only by a chemical destruction at mid-latitudes but also by the advection of ozone depleted air from low- and high- latitudes. In order to investigate factors that affect the ozone trend observed at the midlatitudes, the effects of stratospheric aerosols, bromine species, Quasi Biannual Oscillation (QBO), and Arctic ozone depletion is examined, analyzing the TOMS data, the ECMWF data, and the outputs of chemical transport models. These analyses shows that (1) the effect of the stratospheric aerosol increase due to the Pinatubo eruption in 1991 on the mid-latitude ozone became small by 1997-1999 (2) the effect of bromine species on the ozone destruction at mid-latitudes is 10-20 DU (3) The ozone low region over the Western Pacific in winter is mainly caused by QBO (4) The period of QBO is largely affected by ozone heating (5) QBO has an effect on the mid- and high- latitude ozone amount through the interaction of the meridional circulation that is induced by QBO and the planetary wave activity in the stratosphere (6) The 1997 Arctic polar vortex has an effect of decreasing the ozone concentration outside the vortex through the air mixing between the polar vortex and the mid-latitudes, but the effect is limited near the vortex. The effect is small in the whole mid-altitudes.

A chemistry-climate model run is performed in the period 1980-2004 including all these factors that is important for the mid-latitude ozone. The calculation succeeded in simulating the rapid ozone decrease after the volcanic eruptions, the following gradual ozone recovery in a several years, ozone QBO, and ozone destruction due to bromine species.

(iv) A study on ozone trend in the Northern Hemisphere using a chemical climate model

We investigated the mid- and high-latitude lower stratospheric N_2O distributions in the northern hemisphere in the years with early and late polar vortex breakup time with a CTM that was used in our last year's study and with the zonal and meridional wind velocities nudged towards the ECMWF ERA40 data. A clear difference in the N_2O concentration between the early and late vortex breakup years are found in the winter and late spring, but not found in the summer time, while our analyses in the last year using a CTM where the zonal wind, meridional wind and temperature were nudged showed a difference in the summer N_2O concentration. In addition to the analyses of the N_2O concentration evolution at mid- and high- latitudes in the last year, we also examined N_2O concentration evolution inside the polar vortex. Results show that the effect of the vertical advection on the N_2O concentration in the lower stratosphere inside the polar vortex is small, dominated by the horizontal motions and small-scale motions, which is different from the situation at mid- and high- latitude lower stratosphere, where the vertical advection of N_2O is a dominant process for the N_2O concentration distribution. We also examine year-to-year variation of N_2O concentration in a future atmosphere associated with the ozone concentration variation using

the outputs from a coupled chemistry-climate model (CCM) for the period of 1980-2050.

The following results are highlighted.

(a) Effects of atmospheric sphericity on the stratospheric chemistry and dynamics over Antarctica

The integration of the nudging CTM and the ensemble runs of the CCM were carried out both for the plane parallel radiative transfer (RT) version (the PPA version) and the spherical RT version (the SA version) under the same initial and surface boundary conditions.

The onset of the ozone reduction over Antarctica shifts to earlier dates and the minimum value of the total ozone becomes lower in the SA version is seen both in the nudging CTM and the CCM. The earlier reduction in the ozone mixing ratio in the SA version than that in the PPA version is caused by the earlier increase of the ClO mixing ratio in the SA version, which is initiated by the upward solar radiation flux at the SZAs greater than 90°.

During the ozone-recovery period over Antarctica, the variation and the absolute value of the total ozone in the PPA version and the SA version using the nudging CTM becomes almost the same after the ozone minimum, while in the CCM the total ozone amount of the SA version stays at lower values than those of the PPA version. The behavior of the CCM results cannot be explained by the ozone destruction due to the chlorine catalytic cycles alone.

In the SA version of the AGCM, the diabatic heating around October over Antarctica is smaller than that in the PPA version because of the larger ozone destruction and the lower short-wave radiation absorption by the ozone molecules. This leads to the lower temperature and the larger latitudinal gradient of temperature and hence the delay of the westerly wind deceleration in the SA version of the CCM. This delay of the westerly wind deceleration leads to the delay of the polar vortex break-up, which prevents the transport of the ozone-rich air masses in the lower latitudes into the polar region. This is the main reason for the delayed ozone recovery over Antarctica in the SA version of the CCM.

(b) Future ozone layer prediction using CCSR/NIES Chemical Climate Model

The chemical climate model, CCSR/NIES CCM, with the T42 horizontal resolution (2.8^o by 2.8^o) was developed. The model includes bromine chemistry and heterogeneous reactions on Polar Stratospheric Clouds (PSCs) of Super-cooled Ternary Solution (STS) as well as Nitric Acid Trihydrate (NAT) and ice particles. Radiation process of Schumann-Runge bands, effects of atmospheric sphericity in solar radiation, non-orographic gravity wave effects, and sedimentation velocity corresponding to PSC radius are also included. The model was run imposed by a future projection of greenhouse gases and halogens from the IPCC A1B scenario and Ab scenario of the Beijing Amendments. The results were analyzed and investigated in terms of the recovery of ozone hole in the future atmosphere.

Fig. 1 shows time series of ozone hole area defined by the area where total ozone amount is less than 220 DU in the south of 40^oS. TOMS observation shows appearance of ozone hole at the beginning of 1980s followed by the rapid growth in 1980s, the slower growth in 1990s, and a maximum of ozone hole area around 2000. The CCM result simulates the time evolution very well. In particular the appearance of ozone hole at the beginning of 1980s and a maximum around 2000 are reproduced well. CCM shows Antarctic ozone in the future atmosphere: Ozone hole will not be

reduced until 2020. A clear reduction will be seen after 2020 and ozone hole will be disappeared in the middle of this century. The data around 2050 indicates that ozone hole does not totally disappear, because there is an year-to-year variation in the dynamics of the atmosphere and ozone amount over Antarctica is sensitive to the Antarctic polar vortex condition. Fig. 2 shows time evolution of minimum total ozone in the south of 40°S. A time evolution reversed to that of ozone hole area in Fig. 1 is seen. The minimum total ozone decrease rapidly in the 1980s, more slowly in 1990s, reached a minimum around 2000, then increased and recovered in the middle of this century.

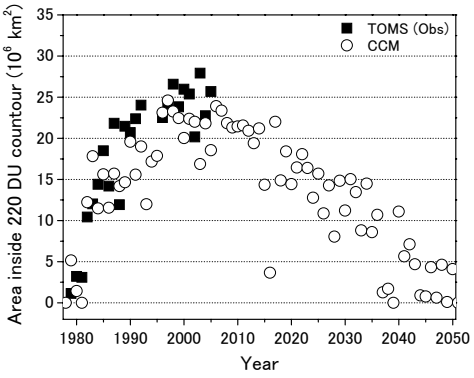

Fig. 1. Time evolution of ozone hole area. Solid squares represent observation by TOMS instrument and open circles represent the results of CCM calculation.

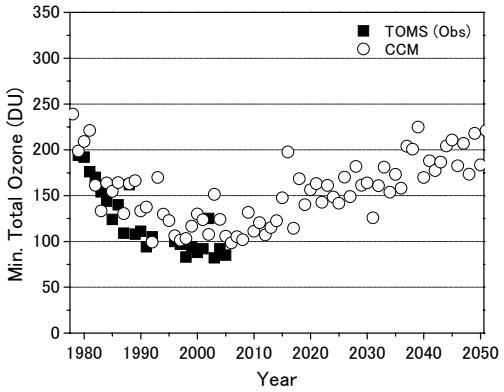

Fig.2. Time evolution of total ozone minimum in the south of 40°S. Solid squares and open circles are same as Fig. 1.

(c) Long-term variation in water vapor around the equatorial lower stratosphere

One of the most highlighted trends in the stratosphere is an increasing one in water vapor, though it is not directly related to the anthropogenic perturbation.[1)] Oltmans and Hofmann[2)] showed that the water vapor concentration in the lower stratosphere was increasing at a rate of about 1 %/year, but this is only based on the water vapor sonde measurements conducted at mid-latitude, Boulder CO, USA. At the tropical tropopause layer where the tropospheric air mass is dehydrated to enter the dry stratosphere, however, we have not had enough observational evidences to indicate the long term variation in water vapor so far. To make this point clear we have been conducting water vapor sonde observations at the equatorial stations in collaboration with the NOAA/ESRL(Earth System Research Laboratory).

Figure 3 shows results from the NOAA water vapor sonde measurements, plotting water vapor concentrations averaged over the 19- 21 km height range. This includes observations during the CEPEX (Central Equatorial Pacific Experiment) campaign in 1993 and those in Brazil done by the NOAA collaborator, and those conducted in our previous campaigns in the eastern Pacific at the Galapagos Islands with the NOAA collaborator. Though water vapor concentrations are highly variable in the equatorial lower stratosphere in accordance with the annual variation of temperature, these sonde observations were mostly done in the northern winter season. The observational results show an increasing tendency in general, though there are several uncertainties such as in the instrumental performance continuation. The trend is estimated as 1 ppmv increase for 14 years,

which is similar to that reported in SPARC (2000).

Randel et al.[3)] showed that the water vapor mixing ratio observed by the satellite decreased after 2001, and that the related temperature decrease is also observed in the tropical tropopause layer. Though the number of observations during 2001- 2004 in Fig. 3 is small, we may see some smaller values at this time. After that we have rather larger values, and may conclude that there is an increasing trend for the last 14 years. We need further investigation for the trend with use of the satellite observations to see the consistency among these datasets.

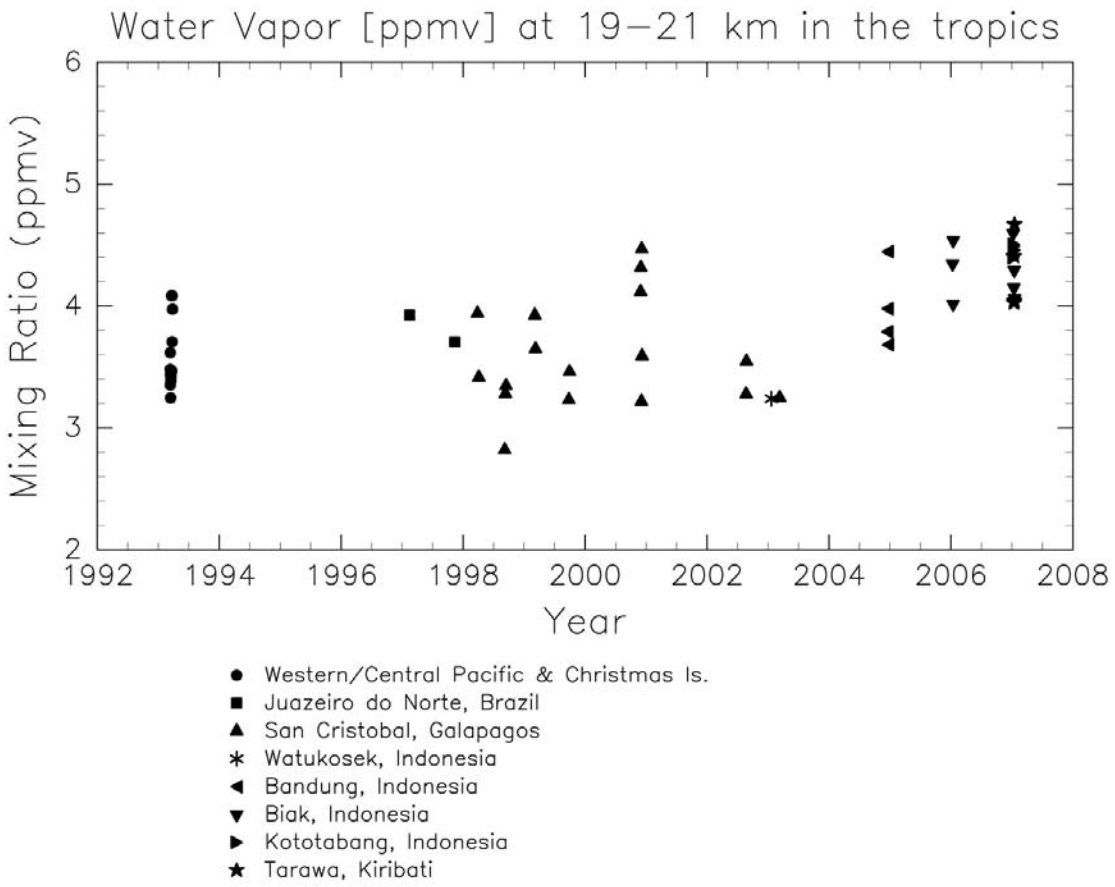

Fig. 3. NOAA water vapor sonde measurements, plotting water vapor concentrations averaged over the 19- 21 km height range

(d) Ozone minimum over the subtropical northwestern Pacific

It is well known that a local total ozone minimum appears over subtropical western Pacific in the Northern Hemisphere in winter. To examine the vertical structure of this ozone minimum, vertically profiles of HALOE ozone data were analyzed. The total ozone minimum was found to be due to the minimum in the stratospheric ozone. It was also found that the vertical profile of ozone mixing ratio deviation from the zonal mean shows bimodal structure: one minimum at around 20 km and the other at around 30km. The analysis with a simple photo-chemical transport model suggested that the mid-stratospheric low ozone is due to southward transport of high-latitude ozone-poor air and the ozone minimum in the lower stratosphere is caused by northward advection of ozone-poor air from the equatorial region.

A nudging Chemical Transport Model (nudging CTM) developed at NIES was applied to simulate the total ozone minimum over subtropical. The horizontal distribution of observed ozone minimum was well simulated by the model. The vertical structure of the ozone mixing ratio deviation from the zonal mean was also calculated using the nudging CTM. The distinctive features of the vertical distribution, i.e., the two negative deviation from the zonal mean, could be reproduced by the model.

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