Summertime Surface N_2O Concentration Observed on Fildes Peninsula Antarctica: Correlation with Total Atmospheric O_3 and Solar Activity

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ABSTRACT

Three-year summertime surface atmospheric N_2O concentrations were observed for the first time on the Fildes Peninsula, maritime Antarctica, and the relationships among the N_2O concentration, total atmospheric O_3 amount, and sunspot number were analyzed. Solar activity had an important effect on surface N_2O concentration and total O_3 amount, and increases of sunspot number were followed by decreases in the N_2O concentration and total O_3 amount. A corresponding relationship exists between the N_2O concentration and total atmospheric O_3, and ozone destruction was preceded by N_2O reduction. We propose that the extended solar activity in the Antarctic summer reduces the stratospheric N_2O by converting it into NO_x, increases the diffusion of N_2O from the troposphere to the stratosphere, decreases the surface atmospheric N_2O, and depletes O_3 via the chemical reaction between O_3 and NO_x. Our observation results are consistent with the theory of solar activity regarding the formation of the Antarctic O_3 hole.

Key words: Fildes Peninsula, Antarctica, nitrous oxide, ozone, sunspot

1. Introduction

Nitrous oxide (N_2O) is a long-living greenhouse gas in the atmosphere. Tropospheric N_2O can be transported to the stratosphere where it produces NO_x, one of the largest contributors to O_3 destruction through different photochemical reactions. This has generated environmental concerns about N_2O because of the increasing UV radiation at the Earth surface due to stratospheric ozone depletion (Crutzen, 1970; Crutzen and Ehhalt, 1977; McElroy and McConnell, 1971). Therefore N_2O emission from sources and its depletion of stratospheric O_3 have been scrutinized closely (Farman et al., 1985; Solomon et al., 1986; Mosier et al., 1991; Dobbie et al., 1999; Fahey et al., 2000; Fahey and Ravishankara, 1999). Since the discovery in 1985 of an ozone hole over Antarctica in the later winter and early spring, intense research efforts have clarified the roles of atmospheric transport and chemistry in the stratospheric ozone changes. The initial focus of this research was on the wintertime conditions, but more recently the stratospheric polar summer has also received a lot of attention (Farman et al., 1985; Tung Ka-kit et al., 1986; Callis and Natarajan, 1986; Solomon et al., 1986; Fahey and Ravishankara, 1999; Bian et al., 2002). The solar-induced effects, such as solar proton events and energetic particle precipitation, have been found to contribute to the ozone depletion in the 11-year solar cycle (Martin et al., 1999).

In this paper, we report the results from three-year summertime measurements of the surface atmospheric N_2O concentrations on the Fildes Peninsula, maritime Antarctica. This is part of the Summer Antarctic Scientific Expedition Project, which was designed to explore the relationships among surface N_2O concentration, total atmospheric O_3, and solar activity.

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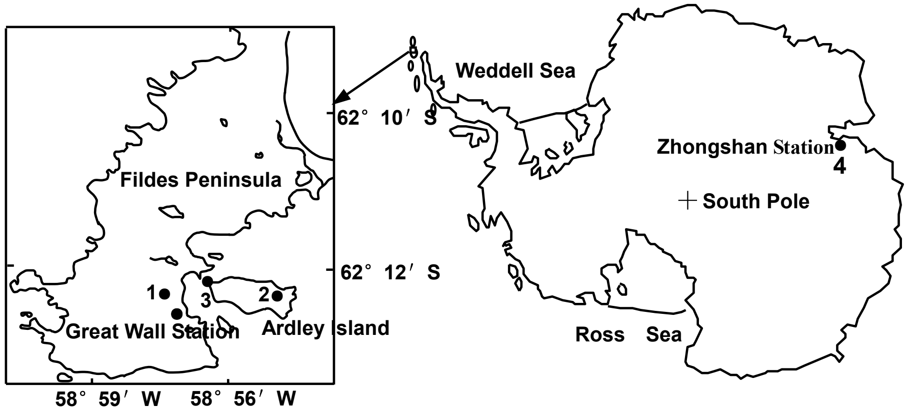

Fig. 1. The observation sites for surface atmospheric N_2O concentration in Antarctica. 1. The tundra observation sites on the Fildes Peninsula; 2. The observation site on Ardley Island; 3. The observation site for Sandy Dam; 4. The observation site at Zhongshan Station.

2. Study areas and methods

2.1 *Study areas*

The gas sampling sites were mainly selected on the Fildes Peninsula in the maritime Antarctic (61°51′–62°15′S, 57°30′–59°00′W), where the Chinese Great Wall Station is located. This peninsula has the characteristics of an oceanic climate, and the predominant wind direction is from the west or northwest because of the effect of the polar cyclone. Meteorological data collected at the Great Wall Station from 1985 to 1995 indicated that the mean annual temperature was about –2.5°C, with a range of –26.6–11.7°C. The annual precipitation, mainly in the form of snow, was 630 mm and evenly distributed throughout the year. In addition, this peninsula is one of the regions with the most research stations. The Russian station Bellingshausen and the Chilean station Teniente Marsh are located on the eastern coast of this peninsula. Ardley Island (62°13′S, 58°56′W), 2.0 km long and 1.5 km wide, is connected to the Fildes Peninsula by sandy dam, and it was defined as an area of special scientific interest by the Scientific Committee of Antarctic Research (SCAR). It is one of the most important penguin colonies in the maritime Antarctic (Sun et al., 2000; Sun et al., 2002). The observation sites for surface atmospheric N_2O concentrations are illustrated in Fig. 1. In summer (January–March) 1999, two observation sites (Sites A and B), about 500 m apart, were set up in the tundra to the northwest of the Great Wall Station. The observation sites were covered by moss (*Bryum Pseudotriquetrum* and *Bryum muelthenbeckii*) and lichens (*Usnea sp*), and were at an elevation of about 41.6m. Since the sites were located upwind (northwest) from the Great Wall Station, little direct impact from the station is expected. In summer 2000, three observation sites were set up in the tundra (Site A), on the sandy dam, and on Ardley Island, respectively. In summer 2002, the same observation site (Site A) was also set up in the tundra on the Fildes Peninsula, and another observation site was set up upwind at the upland near Zhongshan station to avoid impact from the station.

2.2 *Gas sampling and N_2O concentration determination*

We used gas sampling vials (17.5 mL) from the Institute of Japanese Agricultural Environment to collect two duplicate vials of gas samples for each measurement. The average of their final test values of N_2O was used as the surface atmosphere N_2O concentration at the observation site. The vials had been vacuumed to -1.0×10^5 Pa in advance. In the summers of 1999 and 2000, the sampling frequency was once every three or six days and the sampling time was about 12:00 (Chilean time). The sampling duration was 21 January–23 March in summer 1999 and 7 January–23 March in summer 2000. In the summer of 2002, the sampling frequency was once a day in order to examine the relationship between the surface N_2O concentration and total atmospheric O_3 more explicitly. The sampling duration was 25 December 2001–6 March 2002 at the tundra site of Fildes Peninsula and 25 January–30 February 2002 at the observation site near Zhongshan Station. We faced the wind to avoid anthropogenic disturbance during every sampling. The gas samples were randomly collected in the vacuum vials from 2 m above the ground. These vials were sealed with a butyl rubber septum and then covered by a plastic cap. Control experiments showed that the air samples in the vials had minimal and insignificant changes during the sampling, storage, and transporta-

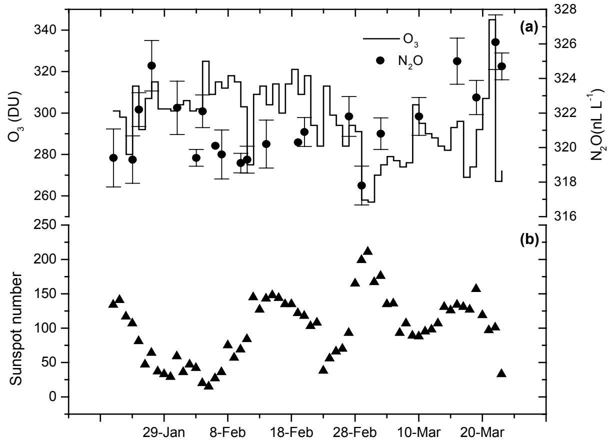

Fig. 2. Surface atmospheric N_2O concentration at tundra observation sites, total atmospheric O_3 on Fildes Peninsula, southwestern Antarctica (a) and sunspot number (b) in summer 1999. Note: average N_2O concentration and standard deviation were calculated according to the data at tundra sites A and B and are shown in the figure.

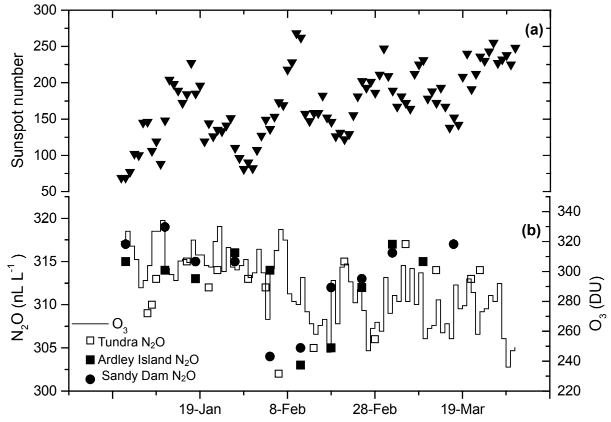

Fig. 3. Surface atmospheric N_2O concentrations at three observation sites, total atmospheric O_3 on the Fildes Peninsula, southwestern Antarctica (b) and sunspot number (a) in summer 2000.

tion period (Sun et al., 2000; Sun et al., 2002). The high vacuum of -1.0×10^5 Pa inside the vial can be maintained for at least one year (Xing, 1998; Xing and Zhu, 1997), and gas standards stored in the vials had no remarkable changes in concentration during 3 months of storage in the laboratory and during transport from the field site to the laboratory.

The collected gas samples were brought back to China and analyzed in the laboratory of Material Cycling in the Pedosphere, Institute of Soil Science, Chinese Academy of Sciences, for N_2O concentrations. The concentration was determined by HP5890 II GC (Gas Chromatograph) using a ^{63}Ni electron capture detector (ECD) (Xing and Zhu, 1997; Sun et al., 2002). The GC-ECD was equipped with a back flush system with 10-port valves. A pre-column (2 m) and a main-column (Porapak Q, 100 mesh) were used with an argon-methane (95%: 5%) mixture as the carrier gas at a flow rate of 30 mL min^{-1}. The detector and column temperatures were 330°C and 85°C, respectively. The back flush time was 2.8 min. The injecting gas volume was 3.0 mL, made by using an adjustable pressure syringe injector. Compressed air was used as a standard gas with the value of 303 nL L^{-1} demarcated by the National Institute of Japanese Agricultural Environment. The variance coefficient for the standard samples was within 0.1%–0.4% in ten hours.

2.3 *Sources for the total atmospheric ozone and sunspot number data*

According to longitudinal and latitudinal values, the data for the total atmospheric O_3 and sunspot number at the observation sites were downloaded from (http://toms.gsfc.nasa.gov/ozone) and (http://www.sec.noaa.gov), respectively. In this paper, the continuous data for total atmospheric O_3 and sunspot number were analyzed during the observation for surface N_2O concentration.

3. Results

The mean N_2O concentration at tundra observation sites A and B on the Fildes Peninsula was 321.4 ± 2.2 nL L^{-1} in the summer of 1999. As seen from Fig. 2, there exists a corresponding relationship between the surface N_2O concentration and total atmospheric O_3. The N_2O concentration increased with the total O_3 amount in the last ten days of January and reached a high value at the end of January, and then fell in the first ten days of February. After that, the N_2O concentration and total O_3 amount reached low values on 12 February at about the same time, and then the N_2O concentration gradually increased before dropping to the lowest value in early March. The total atmospheric O_3 also plunged to its lowest value at the same time. Later in March, the N_2O concentration and total O_3 rose with fluctuations and reached the highest values on 22 March. The N_2O concentration did not completely correspond to the total atmospheric O_3 in February, possibly due to discontinuous sampling and other environmental factors. The sunspot number showed an opposite relationship with the N_2O concentration and total O_3 (Fig. 2). Three peaks of the sunspot number corresponded to three troughs of N_2O concentration and total O_3 quite well, suggesting that the increase in the sunspot number led to the decrease in the N_2O concentration and total atmospheric O_3. Especially in the first ten days of March, the largest O_3 loss and lowest surface N_2O concentration appeared as the strongest solar activity occurred.

In the summer of 2000, the surface atmospheric N_2O concentrations were observed at the tundra, Ardley Island, and Sandy Dam, shown in Fig. 3. The mean concentrations were 310.7±4.4, 312.4±5.1, and 312.7±5.3 nL L^{-1}, respectively. The N_2O concentration profiles at the three sites were consistent (Fig. 3). The time-corresponding relationship between the N_2O concentration and total O_3 was also observed in this summer. The N_2O concentration and total O_3 had insignificant variations from the end of December 1999 to January 2000. After that, the N_2O concentrations at all three sites dropped to their lowest values during the 3–18 February period, indicating that the decrease of the surface atmospheric N_2O concentration was a natural phenomenon in the region of Fildes Peninsula. After a lag of 5–10 days, the total atmospheric O_3 also plunged. In the first ten days of March, the N_2O concentration and total O_3 reached their peak values at almost the same time. The inverse relationship between the sunspot number and the N_2O concentration and total O_3 in Fig. 3 is not as evident as in Fig. 2, but the largest sunspot number occurred during the same 3–18 February period, corresponding to the abrupt drop of the N_2O concentration and total O_3.

To further verify the relationships among the surface N_2O concentration, total atmospheric O_3, and solar activity, we observed the N_2O concentration on every day in the summer of 2002, and the results are given in Fig. 4. The surface N_2O concentration and total O_3 showed quite similar profiles at the tundra observation site on the Fildes Peninsula, southwestern Antarctica (Fig. 4a). The surface N_2O concentration and total atmospheric O_3 increased slowly and reached their highest values during the second set of ten days in January and then gradually decreased. They reached the lowest values around the same time on 10 February and then jumped up to the second peak values during the second set of ten days in February. A significant correlation between the N_2O concentration and total O_3 amount was observed (correlation coefficient $r_1 = 0.56 > r^{70}_{0.01}$=0.30, $r^{70}_{0.01}$ represents the correlation coefficient at the level of 0.01 and with sample number of 70). On the other hand, two peaks of the sunspot number coincided with two troughs of the N_2O concentrations and total atmospheric O_3, implying that solar activity contributed to the drop of the N_2O concentration and total atmospheric O_3 (Fig. 4c). The corresponding relationship between the surface N_2O concentration and total atmospheric O_3 was also observed at the site near Zhongshan station in southeastern Antarctica (Fig. 4b). The N_2O concentrations and total O_3 both had low values from 25 January to 9 February and leaped to the peak values during the second set of ten days of February. The highest N_2O concentration was above 335 nL L^{-1}. The two gases decreased to lower values after 15 February. A significant correlation between the N_2O concentration and total O_3 amount was also observed (correlation coefficient $r_2 = 0.31 > r^{35}_{0.05}$=0.305, $r^{35}_{0.05}$ represents the correlation coefficient at the level of 0.05 and with sample number of 35). The sunspot number was also inversely correlated with the surface N_2O concentration and total O_3 amount at this site. Overall, the summertime variation patterns of the surface N_2O concentration, total O_3 amount, and sunspot number at both observation sites are similar, with the changes near Zhongshan Station slightly ahead of those on Fildes Peninsula.

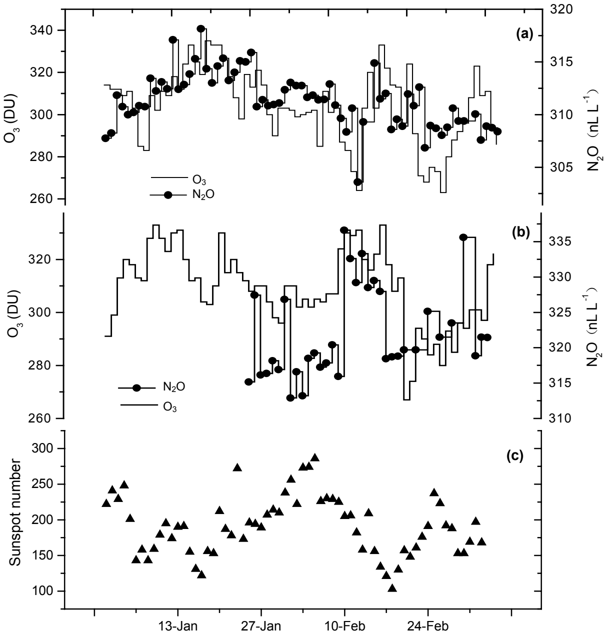

Fig. 4. Surface N_2O concentrations, total atmospheric O_3, and solar activity in summer 2002. (a) N_2O concentrations and total atmospheric O_3 at the tundra observation sites on Fildes Peninsula, southwestern Antarctica; (b) N_2O concentrations and total atmospheric O_3 at the observation site at Zhongshan Station, southeastern Antarctica; (c) Sunspot number.

4. Discussion

4.1 *The time-corresponding relationship between the surface* N_2O *concentration and total atmospheric* O_3

Our observations that abrupt decreases of the surface N_2O concentration were always followed by rapid depletions of ozone suggest the existence of a corresponding relationship between the surface N_2O concentration and total atmospheric O_3. The atmospheric concentration of N_2O in Antarctica depends on the balance between the local emission, air transport, and its sink. Observations indicate that N_2O emissions from the local tundra and aquatic environment are insignificant because of low fluxes and very small emission areas (Sun et al., 2002; Priscu et al., 1996; Priscu, 1997). Therefore the changes of atmospheric N_2O concentrations are more related with the sink strength and air transport in the Antarctic summer. In recent years, studies have shown that atmospheric N_2O cannot be depleted in the troposphere, and its dominant sink is the photolysis and photochemical reactions with O_3 after it is transported from the troposphere to the stratosphere (IPCC, 1992; Crutzen, 1970; McElroy and McConnell, 1971; Farman et al., 1985; Solomon et al., 1986) through the following well studied chemistry processes:

$$N_2O + O \rightarrow 2NO \qquad \text{(R1)}$$

$$NO + O_3 \rightarrow NO_2 + O_2 \qquad \text{(R2)}$$

$$NO_2 + O \rightarrow NO + O_3 \qquad \text{(R1)}$$

Vertical N_2O profiles have been observed at many global sites. Results show that no appreciable vertical gradient of N_2O concentration (311 nL L^{-1}, the concentration generally observed in the troposphere) exists in the troposphere due to rapid atmosphere transport and dispersed distribution of surface sources (Collins et al., 1996; Kondo et al., 1999; Reid et al., 2000; Ishijima et al., 2001; Zhu et al., 2003). In the stratosphere, however, N_2O concentration is generally lower than 250 nL L^{-1} and it decreases rapidly with increasing height due to the destruction of N_2O by the reaction R1, which results in a considerable vertical gradient of the N_2O concentration between the troposphere and stratosphere (Kondo et al., 1999; Collins et al., 1996; Reid et al., 2000). The vertical N_2O concentration profile observed inside the Arctic vortex also showed the same variation (Kondo et al., 1999). Therefore we propose that the N_2O-rich air at the surface and in the lower troposphere can diffuse to the stratosphere because of the concentration gradient. In the stratosphere, N_2O is converted into NO_x (NO and N_2O), and NO_x leads to the depletion of total atmospheric O_3 through the reaction R2. Since air transport and diffusion can be influenced by many environmental factors, the time-corresponding relationship between the surface N_2O concentration and total atmospheric O_3 can be complex and sometimes ambiguous, such as during the first ten days of January in 1999 and 2000.

4.2 *The effects of solar activity*

Our observation results show that solar activity has an important impact on the surface N_2O concentration and total atmospheric O_3. The increase in sunspot number was associated with the reduction in the N_2O concentration and total O_3 amount as shown in Figs. 2–4. The chemistry of the atmospheric N_2O sink and summertime O_3 destruction via the effects of solar activity has been exclusively studied in the polar region (Fahey and Ravishankara, 1999; Fahey et al., 2000; McElroy and McConnell, 1971). O_3 is produced via solar ultraviolet photolysis of oxygen and destroyed through catalytic cycles involving reactive nitrogen (NO_y), halogen (chlorine and bromine), and hydrogen species (HO_x) (Callis and Natarajan, 1986; Solomon et al., 1986). Among the NO_y species, NO_x (NO and NO_2)is the major catalyst. Polar stratospheric high temperatures during the summer preclude the intense heterogeneous activation of chlorine, and NO_x becomes the predominant catalyst for O_3 loss (Portmann et al., 1999). Since large regions of the polar stratosphere receive uninterrupted sunlight for many weeks, it is reasonable to suggest that solar activity reduces N_2O in the stratosphere by converting it into NO_x, and destructs O_3 via NO_x through reactions R1 and R2. As more N_2O is depleted in the stratosphere, the low troposphere N_2O will move and diffuse into the stratosphere more rapidly due to the increase in the concentration gradient. Since O_3 production rates and the transport of ozone-rich air from both lower and higher altitudes are too slow to offset O_3's destruction by NO_x, total O_3 continuously decreases throughout high latitudes in late spring and summer (Fahey and Ravishankara, 1999; Fahey et al., 2000). This model is consistent with our observation results and the theory of solar activity for the formation of the Antarctic O_3 hole.

Solar activity may also explain the observed annual variation of surface N_2O concentration and total atmospheric O_3 in the Antarctic summer. The mean N_2O concentration was about 10 nL L^{-1} higher in the summer of 1999 than in 2000 (Table 1). The N_2O emission from Antarctic local sources is not significant enough to cause that change, and the variation in the global average N_2O concentrations during 1999 and 2000 was minimal (Sun et al., 2002; Priscu et al., 1996; Priscu, 1997). Therefore the inter-annual changes of N_2O concentrations may be related to annual N_2O sink strength. In fact, the sunspot number in 2000 reached the cycle 23 maximum. The sunspot numbers in summer 2000 were more than those in 1999, especially in

Table 1. The comparisons between mean sunspot number, total atmospheric ozone and surface N_2O concentrations in the summers of 1999 and 2000.

Duration	Sunspot Number			Total O_3 (DU)			N_2O (nL L^{-1})		
	Range	Average	n	Range	Average	n	Range	Average	n
Jan–Mar 1999	15–211	98.9±46.5	62	257–345	296.4±17.5	62	317.8–326.1	321.4±2.2	22
Jan–Mar 2000	69–268	168.9±48.9	91	236–334	290.2±22.6	91	302–319	312.8±4.6	36

Note: n represents the number of the samples.

the first ten days of February and the last ten days of March 2000 (Figs. 2–3). High-energy particles produced by solar activity can cause atmospheric ionization, increase odd nitrogen compounds (NO_x), and destroy the Antarctic ozone layer (Zhang and Lu, 1999; Brasseur and Solomon, 1984). Yoshiro (1986) found that sunspot number was negatively correlated with total atmospheric ozone ($R = -0.70$) and the 3-year sliding average of total ozone also exhibited the 11-year cycle. Sunspot activity was greatly intensified in the summer of 2000; it accelerated stratospheric N_2O conversion into odd nitrogen compounds (NO_x), increased the concentration gradient between the troposphere, and stratosphere, and caused more N_2O in the lower troposphere to be transported and diffused to the stratosphere. Therefore mean surface N_2O concentration on the Fildes Peninsula in summer 2000 was abnormally lower than in summer 1999.

5. Conclusions

In summary, the data for three-year summertime surface N_2O concentrations were obtained for the first time on the Fildes Peninsula of maritime Antarctica, and mean concentrations at tundra observation sites were 321.4±2.3 nL L^{-1}, 310.7±4.4 nL L^{-1}, and 311.7±4.5 nL L^{-1}, respectively, in the summers of 1999, 2000, and 2002. Also the three summertime changes of surface N_2O concentrations were correlated with total atmospheric O_3 and solar activity. Analyses of surface N_2O concentrations, TOMS (NASA Total Ozone Mapping Spectrometer) O_3 data, and NOAA (National Oceanic and Atmospheric Administration) sunspot data revealed the following intrinsic relationships:

1) Sunspot number was negatively correlated with surface N_2O concentration and total O_3 amount, and the increases of sunspot number were followed by decreases in N_2O concentration and total O_3 amount;

2) The N_2O concentration and total atmospheric O_3 have a corresponding relationship, and N_2O reductions were followed by ozone destructions.

These relationships, together with the well-studied photochemistry of N_2O and O_3, are consistent with the theory of solar activity about the formation of the Antarctic O_3 hole. Therefore the authors propose that the extended solar activity in the Antarctic summer reduces the stratospheric N_2O by converting it into NO_x, increases the diffusion of N_2O from the troposphere to the stratosphere, decreases the surface atmospheric N_2O, and depletes O_3 via the chemical reaction between O_3 and NO_x.

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