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A primary study of nitrous oxide emission in agriculture region of Northern China

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Abstract—Significance of nitrous oxide in atmospheric chemistry is reviewed briefly. Background concentration of N_2O at Wudaoliang in Qinghai Province of Western China has been measured by collecting gas samples and analysing them with electron capture-gas chromatography. The atmospheric concentration fluctuates in the range of 303-315 ppb with the mean value of 308 ppb. The emission rate of N_2O in agriculture region of Northern China has been studied primarily. Fertilization, rain and temperature of soil appear conductible to N_2O emission. The interactions between greenhouse effect and N_2O emission have been discussed. The catalytic action of N_2O in stratospheric ozone depletion processes and the effect of high N_2O concentration on ozone depletion in stratosphere have been considered and discussed.

Keywords: nitrous oxide; agriculture region; greenhouse effect; Northern China.

INTRODUCTION

The N_2O concentration of the atmosphere has increased from 280–285 ppb 100 years ago to about 310 ppb today. The present rate of increase is estimated about 0.25% per year, corresponding to net emission of 3.5 Tg per year. About 5% of global warming effects are attributable to N_2O, and in spite of its relatively low concentration, the long atmospheric lifetime of this gas (100–200 years) gives it a greater significance in greenhouse effect and in the depletion of stratospheric ozone concentration. It is estimated that of the order of 90% of N_2O emission are from soil and land-use practices, but not all these sources are well known. Reasonable estimates, however, exist of world-wide agriculture imputs from biological and industrial fixation, and the loss of 2–3% of these N (ca 150 Tg/year) as N_2O would account for the entire observed increase in atmospheric concentration. Almost 45% of the world fertilizer consumption currently takes place in North America and Western Europe, but the need to

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increase food production in temperate Asia, especially in China and India, and Eastern Europe has led to increased fertilizer use in these areas. In order to understand the trend of atmospheric concentration and emission rate of N_2O from agriculture land of China, and also its contribution to the depletion of stratospheric ozone and the greenhouse effect a project has been started. Some primary results of this study are shown and discussed in following.

SIGNIFICANCE OF N_2O IN ATMOSPHERIC CHEMISTRY

Arthur Adel discovered atmospheric nitrous oxide (N_2O) in 1938 by using its infrared absorption at 7.77 and 8.57 μm in Solar spectra (Adel, 1939). The strong infrared absorption of N_2O that enabled its early detection in the atmosphere is one of the reasons that it is an important greenhouse gas and chemically active in stratosphere. N_2O is produced in soils by microbial action, from industry, biomass-burning air crafts and so on. Although its growth rate of 0.25–0.3% per year is lower than that of the other greenhouse-gases, CO_2 and so on, it is 100–1000 times more effective as a greenhouse absorber molecule than CO_2 and contributes about 5% of global warming effects. It has a long life time in the atmosphere of more than 100 years and reaches the stratosphere by atmospheric transport and diffusion. In stratosphere N_2O photodissociation at near-UV is the major sink for N_2O and its decomposition leads to production of odd-nitrogen compounds (NO, NO_2) which are homogeneous catalyst for transformation of stratospheric ozone into oxygen.

The photodissociation of N_2O proceeds as

$$N_2O + h\gamma \text{ - - - - - - } \rightarrow N_2 + O('D) < 341\mu m, \quad (1)$$

and the other reactions are N_2O decomposition by O('D)

$$N_2O + O('D) \text{ - - - - - - } \rightarrow 2NO, \quad (2)$$

$$N_2O + O('D) \text{ - - - - - - } \rightarrow N_2 + O_3, \quad (3)$$

The odd-nitrogen compounds have important significance in stratospheric photochemistry proposed by Crutzen (1970) and Johnston (1971). Part of stratospheric ozone is destroyed by catalytic decomposition with odd-nitrogen reaction.

$$NO + O_3 \text{ - - - - } \rightarrow NO_2 + O_2, \quad (4)$$

$$O_3 \text{ - - - - - } \rightarrow O + O_2, \quad (5)$$

$$O + NO_2 \text{ - - - - - - } \rightarrow NO + O_2, \quad (6)$$

.....

$$\text{net}: 2O_3 - - - - \rightarrow 3O_2. \quad (7)$$

within the natural stratosphere, catalytic reactions of NO and N_2O are the most important reagents such as CFC and so on, although human made chloride compounds also have important impact on stratospheric ozone and their emission into the air is growing rapidly.

On the other hand the reactions between odd-nitrogen compounds and ClO_x are also significant for protecting stratospheric ozone against otherwise more rapid destruction by the ClO_x catalysts.

$$ClO^- + NO - - - - \rightarrow Cl^- + NO, \quad (8)$$

$$ClO^- + NO_2 + M - - - - \rightarrow ClONO_2 + M, \quad (9)$$

$$Cl^- + CH_4 - - - - \rightarrow HCl + CH_3. \quad (10)$$

Reaction (9) converts both NO_x and ClO_x catalysts into $ClONO_2$ which does not react with ozone, meanwhile reactions (8) and (10) transform ClO_x into unreactive HCl.

NO_x also reacts as a homogeneous catalyst to destroy OH radicals in the stratosphere

$$OH^- + NO_2 + M - - - - \rightarrow HNO_3 + M, \quad (11)$$

$$OH^- + HNO_3 - - - - \rightarrow H_2O + NO_3^-, \quad (12)$$

$$NO_3^- + h\gamma - - - - \rightarrow NO_2 + O, \quad (13)$$

.....

$$\text{net}: 2OH^- - - \rightarrow 2H_2O + O, \quad (14)$$

Since OH^- can react with HCl to produce Cl^-

$$HCl + OH^- - - \rightarrow Cl^- + H_2O, \quad (15)$$

the consequence of Reaction (14) leads to ozone protection as it decreases the production of reactive Cl^- from HCl in Reaction (15).

BACKGROUND CONCENTRATION AND EMISSION RATE OF N_2O

Although the background concentration and emission rate of N_2O have been studied in many regions and places of the world for relatively long time (Weiss, 1981; Elkins, 1989; Prinn, 1990), its background concentration and emission rate on Asian continent and Chinese agriculture regions have not been studied before. There are varieties of N_2O sources in China: fossil fuel burning, biomass burning, oceans, soils, plants and so on. The emission of N_2O from agriculture sources is thought to be the most important and large one, and also is one

of the very complicated, important and still unclear factors for global changing study. Regarding to the N_2O agriculture source in China, it is more complicated since in the last forty years (1949–1989), the population has become 1.8 times higher, and consumption of chemical fertilizer—more than 100 times higher, so the contribution of N_2O from Chinese agriculture regions is an important problem to be studied for estimating climatic change.

On the Chinese continent, the background and regional concentration of atmospheric N_2O only began to be measured recently (Su, 1989). At Wudaoliang in Western China atmospheric background of trace gases has been studied. The location of the sampling station for background studying is 36°25′N, 94°50′E, 4300 m above sea level (Fig. 1). Collected samples have been analysed with electron capture-gas chromatography (ECD-GC). The range of N_2O concentration is 303–315 ppb, with a mean value of 308 ppb.

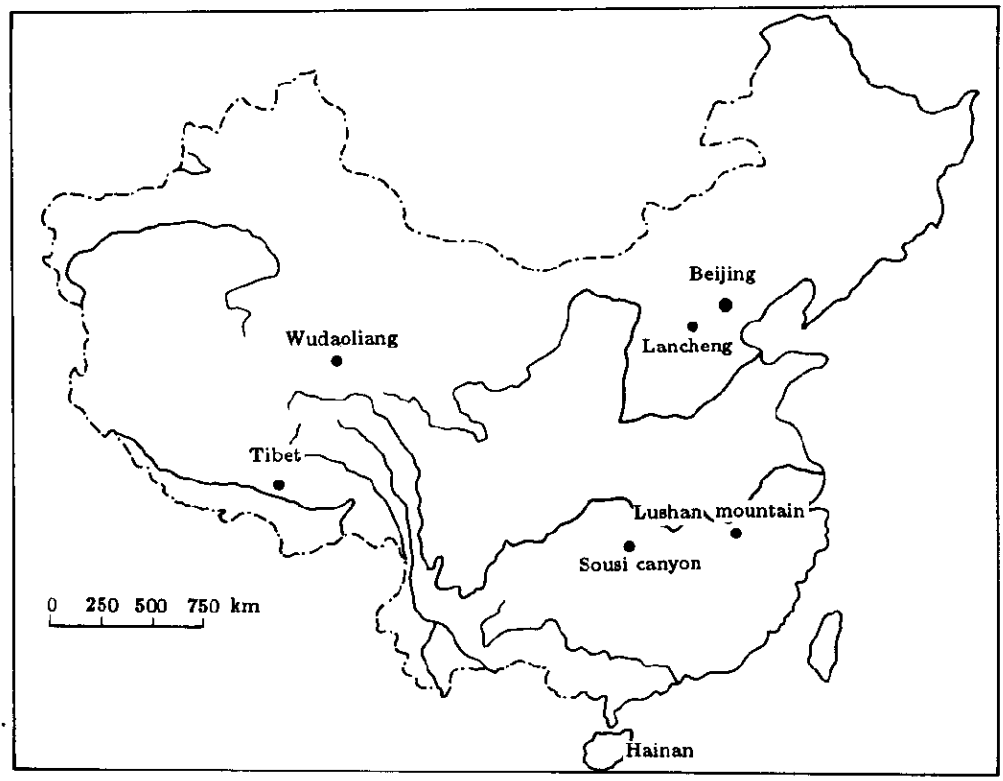

Fig. 1 Places of sampling in China

For measurement of N_2O concentration in agricultural region samples were collected at a vegetable field near Beijing and winter-wheat field in Northern China (37°54′N, 114°42′E) (Fig.

1). The regional concentrations of N_2O are different in vegetable field and winter-wheat field depending on farming processes. The results are shown in Fig. 2.

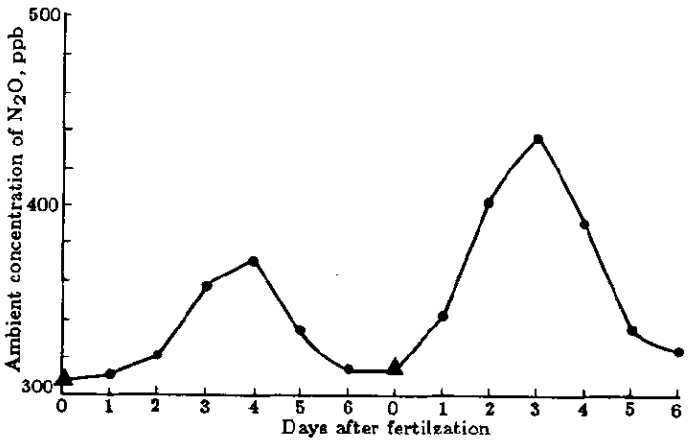

Fig. 2 Ambient concentration of N_2O after fertilization at a vegetable field
△ — the day fertilizer applied

The concentration of N_2O at these agriculture lands is higher than that at background stations in clean area. The farming processes, i.e. rainfall, fertilization and so on, affect the ambient concentration of N_2O. At vegetable field the ambient N_2O concentration rose rapidly to a high peak on the third or forth day following fertilization then fell sharply as shown in Fig. 2. Above the vegetable and winter-wheat fields the ambient concentrations of N_2O have a large range of variation depending on farming practices and meteorological conditions, and most of them are higher than 320 ppb.

The measurement of N_2O fluxes in agriculture region have been performed in vegetable field and winter-wheat field in Northern China by chamber method. In general, high fluxes occurred when soil temperature was high and after the rainfall or irrigation following application of nitrogen fertilizer. For one experiment in an agriculture field the soil temperature at 5 cm bellow surface was 27°C, small rain fall on the day of fertilization and the field was irrigated after 5 days later, as illustrated in Fig. 3, the N_2O emission rate rose to 15 $\mu gN/m^2$. hr on the day of fertilization and light rain, then increased to 25 $\mu gN/m^2$. hr on the day before irrigation. After irrigation the N_2O flux rose steeply to 45 $\mu gN/m^2$. hr, then sharply fell to 5 $\mu gN/m^2$. hr on the 9th day after fertilization as the soil dried.

It was also found in field experiment that the emission rate of N_2O depends on soil tem-

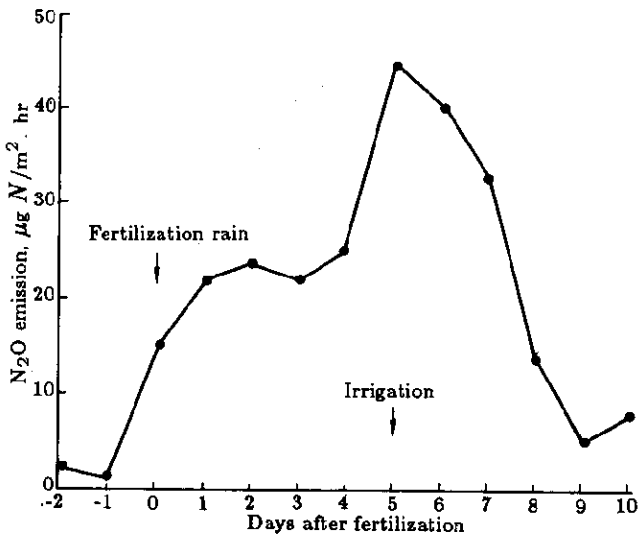

Fig. 3 Change of N_2O flux in agriculture region of Northern China

perature, and soil temperature is conductible to emission. Generally the amplitude of emission variation is greater at higher temperature. But it is difficult to estimate the effect quantitatively on our limited data.

From our primary work, the mean value of N_2O emission rate is in a range of 6–45 $\mu gN/m^2$. hr with a mean value of 15 $\mu gN/m^2$. hr or 0.54–3.6 kgN/ha. year and 1.35 kgN/ha. year respectively.

On basis of data in Table 1 from literature (Bauwman, 1989) and this work, the contribution of N_2O from Chinese agriculture region could be estimated. Provided the area of Chinese agriculture region is about 170 million ha and N_2O flux is probably 1.4 kgN/ha.year. N_2O from Chinese agriculture region is in a range of 0.1–0.61 TgN/Year with a mean value of 0.27 TgN/year. These figures may be acceptable since N_2O emission from fertilized soil in the whole would is as large as 1.5–3 TgN/year (Bauwman 1989) from the cultivated global area of 1500 million ha, although the population in China is about one fifth of the whole population in the would.

Table 1 N_2O emission rates from some croplands

Crop	Fertilizer	Fertilizer application, kgN/ha.	Mean flux range, $\mu gN/m^2$. h		Annual Sources emission range , kgN/ha. a		
Winterwheat	NH_4NO_3	210	19	23	1.7	2.0	Colbourn, *et al*, 1984
Corn	NH_4NO_3	132	25	33	2.2	2.9	Duxbury,
Corn	Organic	130	27	43	2.4	3.8	*et al*, 1982
Barley	NH_4NO_3	56		25		2.2	Mosier,
Barley	Organic	71		29		2.5	*et al*, 1982.
Barley		0		14		1.2	
Barley	NH_4NO_3	70/140	10	64	0.9	5.6	Burford, *et al*, 1981
Alfalfa		0	26	48	2.3	4.2	Duxbury, *et al*, 1982

TREND OF N_2O CONCENTRATION CHANGE AND ITS EFFECT ON ENVIRONMENT

Atmospheric N_2O has its main source from soil (McElroy, 1977; Cicerone, 1978). Pearman *et al.* (1986) found that the per-industrial value of atmospheric N_2O was 268±6 ppb for the period between 1600 and 1800 A. D. from ice cores in Antarctica. Using a linear regression on the whole record of N_2O measurements, the calculate showed that the atmospheric N_2O has increased 6 ppb per century over the past 300 years. In the last decade the average growth rate of N_2O is about 0.63–0.86 ppb (or 0.2–0.25%) per year from flask collected samples at NOAA's Geophysical Monitoring for Climatic Change baseline stations with a background concentration from 300 ppb to 308 ppb (Fig. 4) (GMCC Summary Report, 1987; Weiss, 1981; Elkins, 1989). Recently, Prinn *et al.* (1990) reported that the last decade of atmospheric N_2O measurements from the Global Atmospheric Gas Experiment sites using in situ GC analysis produced a growth rate of about 1 ppb (or 0.3%) per year. The human contribution is dominated by the use of manufactured fertilizers whereby fixed nitrogen applied to the soil partially is reduced to N_2O through denitrification and nitrification mechanism.

This observed rate of N_2O concentration increase represents an atmospheric growth rate of above 3 to 4.5 TgN/year, although it appears not too much comparing to the N_2O atmospheric reservoir of about 1500 TgN. But since the life time of N_2O is long, its effects on environment appears more important. If atmospheric N_2O is to build up significantly, its photodecomposition in the stratosphere will generate excess NO as mentioned above.

One of the environmental effects of N_2O is the greenhouse effect on climate change and

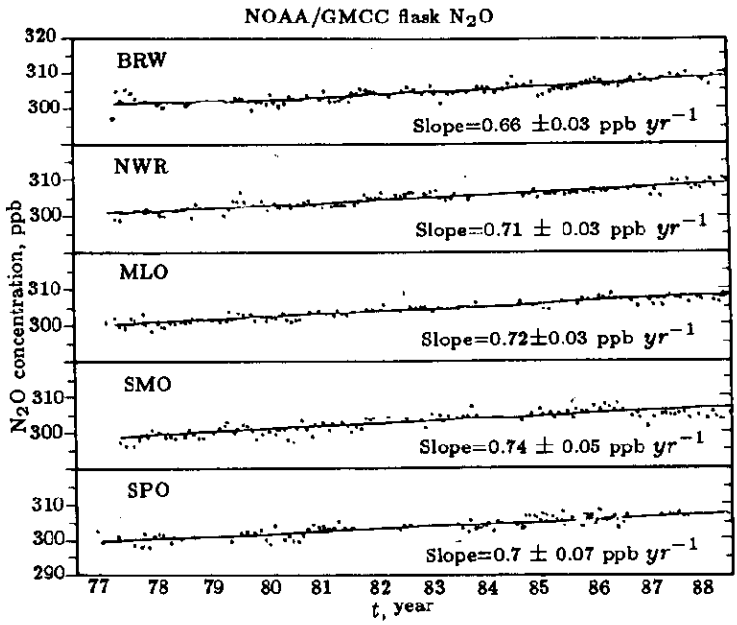

Fig. 4 Atmospheric measurements of nitrous oxide from the NOAA/GMCC network

the feedback action of climate on N_2O emission. Regarding the greenhouse effect the radiative absorption potential of N_2O is about 100–1000 times higher than that of CO_2 (Bauwman, 1989), although the concentration of N_2O is lower than that of CO_2 in 1000 times. Considering the long term changes of atmospheric concentration of N_2O and climate the interaction of N_2O emission from soil and climate is being studied. Investigation showed that higher soil temperature could have some effect to make the N_2O emission rate increased. So if the soil and atmospheric temperature became higher as a consequence of greenhouse effect of trace gases, the emission rate of N_2O from soil could increase to some extent, by turns, N_2O could contribute more to the greenhouse effect. Such interaction between N_2O emission and greenhouse effect needs to be studied further.

The depletion of stratospheric ozone caused by its catalytic conversion into oxygen by odd nitrogen oxides is another effect of N_2O on the environment. Within the natural stratosphere N_2O photodissociates into odd nitrogen oxides, then homogeneous catalytic reactions of NO and NO_2 with ozone, as mentioned above, are the most important ones of the ozone balance. For an assumed doubling of N_2O concentrations (from about 300 ppb to 600 ppb), current

models predict a possible 10–16% ozone depletion (steady-state). The changes in the ozone profile computed at different latitudes in a 2-D model (Whitten, 1983) is illustrated in Fig. 5. The effect of N_2O on atmospheric ozone obviously depends on the altitude and latitude. That also reflects in the altitude and latitude dependence of ozone depletion and in the larger ozone decrease at high stratosphere and higher latitudes.

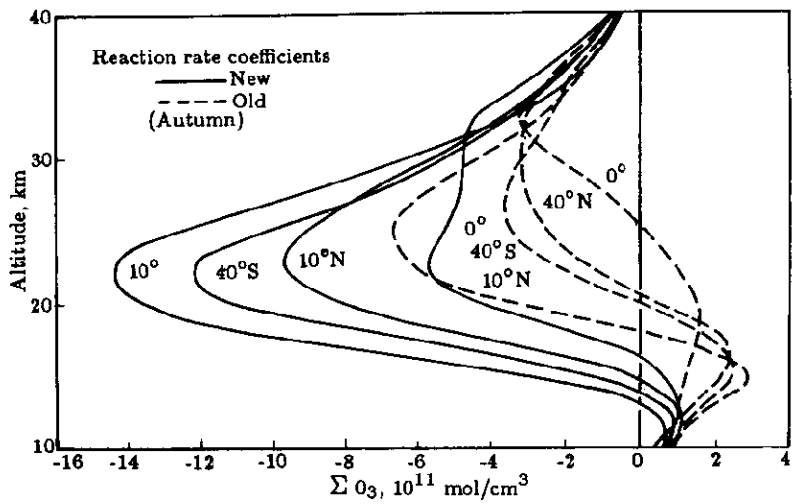

Fig. 5 Predicted steady-state changes in stratospheric ozone concentrations at several latitudes when the atmospheric N_2O abundance is doubled (Whitten, 1983)

CONCLUSION

Nitrous oxide is an important atmospheric trace gas, because it is 100—1000 time more effective as a greenhouse absorber per molecule than CO_2 in spite of its lower atmospheric growth rate than that of CO_2, and unlike CO_2, it also is involved in the depletion of atmospheric ozone. Much of the information available suggests that human activities are responsible for its atmospheric growth and about 90% of N_2O emission are from soil and land use practices. The background concentration and emission rate of N_2O in China have begun to be studied primarily. More research work is necessary for studying the mechanism of N_2O emission, the contribution from different ecological regions, the interaction between N_2O emission and greenhouse effect, and the effect of N_2O emission on U. V. intensity on earth surface through stratospheric ozone depletion and also the ecological consequences.

REFERENCES

Adel, A., Astrophys. J., 1939, 90:627

Bauwman, A. F., The role of soils and land use in the greenhouse effect, ISRIC, Wageningen, Netherlands, 1989

Burford, J. R., R. J. Dowdell and R. Cress, Journal of the Science of Food and Agriculture, 1981, 32:219

Cicerone, R. J., J. D. Shetter, D. H. Stedman, T. J. Kelly and S. C. Llu, J. Geophys, Res., 1987, 83:3042

Crutzen, P. J., Q. J. R. Meteorol. Soc., 1970, 96: 320

Colbourn, P., M. M Iqbal and I. W. Harper, Journal of Soil Science, 1984, 35:11

Duxbury, S. M., D. R. Bauldin, R.E.Terry and R.L.Tate, Nature, 1983, 298: 462

Elkins, J. W. and R.Rosson, Summary Report, 17: Geophysical monitoring for climatic change, NOAA ERL, Bouldea, CO, 1988

Johnston, H.S., Science, 1971, 173:517

McElroy, M.B., S.C.Wofsy and Y.L. Yung, The nitrogen cycle: Perturbations due to man and their impact on atmospheric N_2O and 0.3, R. Soc. London Philos, Trans:, Ser: B, 1977, 277:159

Mosier, A.R., G.L.Hutchinson, B.R.Sabey and J.Baster, Journal of Environmental Quality, 1982, 11:78

Pearman, G.I., D. Etheridge, F. Desilva and P.J.Fraser, Nature, 1986, 320:248

Prinn, R.D. Cunnold, R. Rusmussen, P.Simmonds, F. Alyea, A. Crawforrd, P.Fraser and R.Rosen, 1990: Atmospheric trends and emissions of nitrous oxide deducted from ten years of ALE-GAGE data

Su, W. H., Study of CH_4 and N_2O emission on some stations of eco-environmental measurement and experiment network, paper on International Conference on "Soils and the Greenhouse Effect", Wagenigen, Netherlands, 1989

Weiss, R.F., J. Geophys, 1981, 86 (C8): 7185

Whitten, R. C., W. J. Boruchi, H.T. Woodward, L.A. Capone and C. A. Riegal, Atmos. Env., 1983, 17:1995