A Review of Research on Human Activity Induced Climate Change I. Greenhouse Gases and Aerosols

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ABSTRACT

Extensive research on the sources and sinks of greenhouse gases, carbon cycle modeling, and the characterization of atmospheric aerosols has been carried out in China during the last 10 years or so. This paper presents the major achievements in the fields of emissions of greenhouse gases from agricultural lands, carbon cycle modeling, the characterization of Asian mineral dust, source identification of the precursors of the tropospheric ozone, and observations of the concentrations of atmospheric organic compounds. Special, more detailed information on the emissions of methane from rice fields and the physical and chemical characteristics of mineral aerosols are presented.

Key words: greenhouse gases, aerosol, dust, ozone

1. Introduction

Among the many effects of human activities on climate change, the global scale changes of atmospheric composition due to emissions of industrial activities are the most significant and obvious facts. Recent measurements show that the concentration of atmospheric CO₂ has increased from 280 ppm before the industrial era to 367 ppm in 1999; that of atmospheric CH₄ has increased from 0.8 ppm before the industrial era to 1.75 ppm in 1998; that of N₂O has increased from 0.28 ppm before the industrial era to 0.33 ppm in 1998. At least part of these observed changes may be attributed to human activities (IPCC, 2001). It is generally accepted that the observed changes of atmospheric concentrations of greenhouse gases and aerosols are important causes for the observed climate changes and may further modify the climate in the future. Therefore, study on the causes of these observed changes of atmospheric concentrations of greenhouse gases and aerosols and the prediction of their future trends are of fundamental importance. During the last 5 years or so, China has carried out much research in these fields. This paper summarizes the major results.

2. Researches on greenhouse gas emissions

2.1 Automatic observational system of greenhouse gas emission

An automatic sampling and analyzing system for

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methane emission fluxes, which was designed and assembled by our group on the basis of static sampling and a gas chromatography (GC)-flame ionization detector (FID) analysis technique (Wang, 2001), was employed for continuous measurement in a rice field. Furthermore, we also developed automatic sampling and analysis systems for N₂O (Wang et al., 1997) and NO (Zheng et al., 1998) emission. Using this system, the emission of CH₄, N₂O, and NO from cropland can be simultaneously measured. This system can simultaneously measure field emission data from 16–32 different sampling chamber, 8-12 times per day. It is a useful tool to study the diurnal variation of methane emission in different fields. Since 1985, we have conducted continuous measurements of methane emission in the major rice cultivation regions in China and obtained many valuable methane emission data.

2.2 Mechanisms for the production, oxidation, transportation of CH₄ in rice fields

Methane emission rates from Chinese rice fields have been measured in all five major rice cultivation regions in China (Wang et al., 1998). Four types of diurnal variations of $\mathrm{CH_{4}emission}$ rates have been found (Wang, 2001). Seasonal variation patterns of $\mathrm{CH_{4}emission}$ differ slightly in different field locations, where climate system, cropping system, and other factors are different. $\mathrm{CH_{4}}$ production mainly occurs in the reduced soil layer (2–20 cm). $\mathrm{CH_{4}}$ is oxidized

mainly in the thin surface layer of paddy soi1 and in the rhizosphere of rice plants. Production and oxidation rates are affected by many factors. CH₄ transport occurs in three ways: through the rice plant, gas bubbles and diffusion in flood water. The relative importance of each route is different for different stages during the growing of the rice. The effects of various mineral fertilizers on CH₄ emission are rather contradictory, while the amount of organic manure will enhance CH₄ emission from rice fields, which has also been indicated by CH₄ production rates. Application of fermented sludge from biogas generators and farmyard-stored manure instead of fresh organic manure seems to be promising.

2.3 Model study of methane emission from rice paddies

Developing a methane emission model is an effective approach to accurately estimate regional and global methane emissions. It is also a useful tool to develop mitigation methods of methane emission from rice fields. At present, the numerical modeling is still in a preliminary stage. The first model was developed in 1995 (Cao et al., 1995). Based on 13-year field experiments and studies on methane emission from rice fields, we developed the first version of the methane emission model in China (Ding and Wang, 1996). This model was tested by our experimental data, and was proved able to simulate diurnal variations of CH₄ emission and estimate the total emission of methane from rice fields. In recent years, the methane emission model has seen some new progress (Huang et al., 1998). We are improving our model in order to correctly describe the effects of environmental factors, such as climate, soil properties, fertilizer application, water regime and rice cultivars on processes of methane production, transportation, and oxidation, and precisely predict the methane emission variations under ongoing climatic changes. Using this model, the mitigation options of methane emission from rice fields can be evaluated and valuable mitigation methods can be provided to the government (Li et al., 2002).

2.4 Methane emissions from China's and the globe's rice fields

Based on the 13-year field experiments in all five typical rice fields and model simulation results, China's rice fields contribute 5–13 Tg yr⁻¹ to the atmosphere (Li et al., 2002), much less than the estimates of 30–50 Tg yr⁻¹ made previously by other studies (Khali1 et al., 1991). If we extrapolate the measured data in China with consideration of measured data in other countries, the total global emission of CH₄ from rice is estimated to be 20–40 Tg yr⁻¹ (Wang and Shangguan, 1996). This value has been gradually

accepted and cited. In the new report of the Intergovernmental Panel on Climate Change (IPCC), the methane emission from global rice fields has changed from the 1990s estimated value of 110 Tg yr^{-1} (IPCC, 1990) to 60 Tg yr^{-1} (IPCC, 1995).

2.5 Mitigation technologies of methane emission from rice fields

Based on the 13-year field experiments, we have found that agriculture management is one of the most important factors in determining methane emission from rice fields. Appropriate and scientific readjustment of agriculture management can reduce methane emission without lowering rice yield. Particularly, intermittent flooding irrigation and the usage of biogas residues instead of fresh organic fertilizer are the most worthy techniques in reducing methane emission (Li et al., 1998).

2.6 Emission characteristics of N₂O and NO in croplands

Based on a four-year in situ measurement of nitrous oxide (N_2O) emission from a rice-wheat rotation ecosystem of Southeast China and simulated experiments in the laboratory, the impact of soil moisture on N₂O emission was investigated. All the factors that influence the nitrification and denitrification processes may affect N₂O emission from croplands (Batjes and Bridges, 1992; Williams et al., 1992). These factors include temperature, soil moisture, carbon and nitrogen availability, soil properties, plant growth, microbe activities, chemical and physical disturbances in agricultural operation, and the local climate. Soil moisture directly regulates oxygen availability in soil pores, which determines the biochemical equilibrium status of nitrification and denitrification and the ratios of N₂O to final products. Meanwhile, soil moisture directly influences the processes of N₂O transport and emission from production sites inside soils to the atmosphere. So soil moisture is the most sensitive factor to regulate N₂O emission from croplands. Optimum emission of N₂O from the rice-based agroecosystem was found to occur at the soil moisture level of 99\% water-filled pore space (WFPS), which is different from the 75% WFPS values of soil moisture in grassland and forest (Zheng et al., 1996).

By analyzing the experimental data, two typical patterns of NO diurnal emission from wheat fields were discovered. They are the day-peak pattern and night-peak pattern. For the former, the maximum emission takes place in the early afternoon and the emission at night keeps a relatively low but stable level. This day-peak pattern is closely related to the temperature variation. For the night-peak pattern, the maximum emission occurs during the period of 1800–2400 LST

while the minimum emission occurs in the early afternoon. The intensive up-take of available N (NH $^{4+}$ -N) via wheat plants reduces the NH $^{4+}$ -N supply for nitrification microbes; as a result, less NO is emitted. This principle of competition for available N between plant roots and soil microbes determines the diurnal pattern of NO emission.

Temperature, fertilization, and water status are the key regulating factors of N_2O and NO emissions from the rice-wheat croplands. Reducing mineral N input, wide application of dry fermented organic manure, and sound water management may be the effective mea-

sures to mitigate the emissions of N_2O and NO from croplands.

3. Global carbon cycle

At present, the unbalanced carbon budget (a large "missing" carbon sink) is a hotspot in carbon research. Analysis results for the 1980s indicate that the "missing sink" is about $1.3 \,\mathrm{Pg}\,\mathrm{C}\,\mathrm{yr}^{-1}$ (Woodwell et al., 1998; Schimel, 1998) (see the following equation, units: Pg C yr⁻¹, where $1 \,\mathrm{Pg} = 1 \,\mathrm{Gt} = 10^{15}\,\mathrm{g}$).

Sources Sinks Fossil +Land-use=Atmospheric increase +Northern forests +Oceans +Residual sink $5.5(\pm 0.5)+1.6(\pm 0.7)=3.2(\pm 0.2)$ +0.6(\pm 0.6) +2.0(\pm 0.8)+1.3(\pm 1.1)

Balancing of the carbon sources and sinks becomes the critical problem in the fields of climate and biogeochemistry (Schimel, 1998), and it is an important prerequisite in predicting the future climate. Although there are many "proofs" suggesting that the terrestrial ecosystem absorbs more carbon from the atmosphere (Tans et al., 1990; Cao and Woodward, 1998), direct measurement data are still scarce and sparse. Much research has indicated that ocean plays an important role (Tans and White, 1998). So, ocean and land ecosystems are two key factors needing more investigation.

3.1 The relationship between terrestrial carbon fluxes and climate

Increasing atmospheric CO₂ concentration is the most important factor influencing climatic change which is mainly due to human activities. It is believed that climate changes, carbon, and nitrogen fertilization effects are the three main factors affecting net carbon fluxes of the land ecosystem. At present, more research has focused on the climate effect (Woodwell et al., 1998), as warming would induce more CO₂ release from some soil types and this would enlarge the "missing sink" to some extent. The observed data in the past decades show that the natural atmospheric CO₂ concentration is positively correlated with the global land temperature and negatively correlated with the global land precipitation on a decadal scale (Keeling et al., 1995). By contrast, based on the analysis of observed data, we have found a significant positively correlation between the interannual variability of CO₂ growth rate and the year-to-year changes in the global land precipitation (Yang and Wang, 2000). Thus, a new relationship between atmospheric CO₂ and climatic variables is proposed.

This positive relationship indicates that an increase in current year rainfall does not cause net increases in the absorbed carbon by the vegetation. However,

the increases in total cloudiness will reduce the uptake of carbon by plants. An additional analysis shows that the low latitude regions (especially the Eastern Asia monsoon region) plays a main role in causing this phenomenon. The cloud is the main limiting factor for vegetation growth in the low latitude regions. The increases in cloudiness in low latitude areas would certainly reduce the solar radiation reaching the plant canopy, and consequently lower the photosynthesis rate and cause a positive anomaly in CO₂ emission from the terrestrial biosphere. This mechanism can provide a better explanation for the observed fact first reported by Bacastow in 1976 that the CO₂ flux anomalies lead changes in the Southern Oscillation Index (SOI) by about half a year. Although further investigation is needed, this new finding has an important meaning for the global carbon cycle, especially for the interannual CO₂ variations. Furthermore, this phenomenon closely relates to the climate changes with the global atmospheric CO₂ concentration. So, the effects of clouds (concurrent with precipitation) on solar radiation and then on photosynthesis should be considered in future ecosystem models in order to give a precise description of the ecosystem carbon cycle.

Some researchers applied soil samples obtained from the second soil census in China in 1998 to analyze the correlations of soil organic carbon storage (SOC) and climatic factors, including annual mean temperature (T) and total precipitation (P) (Zhou et al., 2003). The results show that the correlations are quite different in different temperature zones. In the area where $T \leq 10^{\circ}\mathrm{C}$, the correlation between SOC and temperature has the strangest negative correlation. In the area where $10^{\circ}\mathrm{C} < T \leq 20^{\circ}\mathrm{C}$, a positive correlation appears under the influence of precipitation. In the area where $T > 20^{\circ}\mathrm{C}$, the correlation between SOC

and temperature and precipitation is inadequate.

3.2 Model study on ocean carbon cycle

The oceans contain about 40 000 Gt C in dissolved, particulate, and living forms. By contrast, land biota, soils, and detritus contain a total of about 2200 Gt C. Living and dead biogenic matter in the ocean contains at least 700 Gt C, almost equal to the amount of CO₂ in the atmosphere (about 750 Gt C). So, the ocean carbon cycle has an important influence on the global carbon cycle. Marine biota plays an important role in marine biogeochemical processes and in determining the marine carbon cycle. Due to the difficulties in directly collecting marine data, models are useful tools to study marine biogeochemical processes (Maier-Reimer and Hasselmann, 1987; Bacastow and Maier-Reimer, 1990).

A two-dimensional atmospheric CO_2 -ocean carbon cycle model has been developed and used to simulate the surface distributions, vertical distributions, and the isogram distributions along the meridian of various chemical species in the Atlantic (Dong et al., 1994) and Indian Oceans (Pu and Wang, 2000). Based on a two-dimensional ocean thermohaline circulation carbon cycle model, we studied carbon cycling in the Pacific Ocean (Xu and Wang, 1998).

Our models have overcome the shortcoming of the box model and include processes such as the exchange of carbon dioxide between the atmosphere and ocean, photosynthesis, decomposition of organic matter, calcium carbonate production and dissolution, and sinking of suspended particles. In particular, the effect of ocean biota on the carbon cycle is coupled into the model.

The horizontal and vertical distributions of various chemical species in the Indian Ocean have been studied (Pu and Wang, 2000). It was found that the distributions of total carbon both in the ocean and in the atmosphere at a steady state depend on the various chemical and physical processes, and also on the boundary conditions. The distributions of chemical compounds are sensitive to the horizontal diffusion coefficient and photochemical action constant rate. Simulated distributions of the total dissolved inorganic carbon, alkalinity, nutrients, dissolved oxygen, and ¹⁴C isotopes are close to the observed data. It was also found that the ¹⁴C was exported to the deep ocean mainly through a key region in the southern Indian Ocean (10°-30°S), indicating that anthropogenic CO₂ will be transported from the ocean surface to the deep ocean through that region.

4. Asian dust

Dust is a main component of atmospheric aerosols. It is estimated that 1000–3000 Megatons of mineral

dust are injected into the atmosphere every year, accounting for half of the total tropospheric aerosol. The mineral dust particles originate mainly from the great desert areas in northern Africa, southwestern America, and Asia. Mineral aerosols play key roles in atmospheric chemistry, ecology, and the earth's radiative balance. Dust storms not only affects on the air quality, traffic, etc., and cause soil erosion and endanger the lives of human beings and domestic animals, but as a special atmospheric aerosol, dust particles also have significant climate and environment effects. The interactions of dust with earth's radiation field are more complicated than those of other atmospheric aerosols because mineral particles can scatter and absorb solar radiation, leading to either cooling or heating of the climate system under various conditions. Currently, there are large uncertainties regarding the direct radiative forcing from dust, and even larger uncertainties regarding its indirect effects (IPCC 1996; Wang, 2000).

Nowadays, extensive research has focused on dust in the Sahara, but little has been done on Asia dust. Therefore comprehensive study should be carried out on Asian-dust characteristics, influences, and transport mechanisms, etc. Due to the lack of observation data, the parameters in some models are too simple and mostly based on the results obtained in the Sahara desert, so they cannot satisfactorily describe the process of Asian dust formation and transportation. In recent years, Chinese scientists began to study dust storms by such methods as information and laboratory analysis as well as field observation combined with numerical model research. The Chinese Academy of Sciences (CAS) is launching a project called the "International Dust Storm Program". It aims to explain the reason why dust storms occur in Asia, the area they cover, how they travel, and their effect on the human environment.

Statistics show that during the past 49 years, the highest frequency of severe dust storms occurred in the 1950s and the lowest was in the 1990s with a general decreasing tendency, but during 2000–2002 the trend relatively increased (Zhou and Zhang, 2003). In the 1960s, for example, dust storms occurred as often as 80 times a year, while the number went down to less than 40 in the 1980s, when the severity of the storms also decreased sharply. The reason for the sharp decrease, followed by the recent, equally sudden increase is still far from clear, although various theories have been put forward. One is that global warming might play an important role in the downward trend. Warmer conditions in the high latitudes result in fewer cold air invasions, which are closely related to the occurrence of dust storms. But this downward trend was broken in the last three years, when strong cold air movements,

caused by spring north westerlies, triggered much severer sand storms than had occurred in previous years.

In spring 2000, dust storm events occurred frequently in northern China. An analysis of the chemical composition of the super dust storm on 6 April shows that the pollution caused by the dust storm was very serious (Zhang et al., 2000). In the dust storm period, the total concentration of 20 elements reached $1536 \ \mu \mathrm{g \ m^{-3}}$, which is 31.4 times that in spring 1999. Even after the dust storm, the concentration of 20 elements remained at 338.7 $\mu g \text{ m}^{-3}$, which is 7 times that in spring 1999. It is found that the higher peaks of concentration of most element particles appear with diameters above 16 μ m, which is not observed at other times. The number concentrations of coarse particles $(D > 2 \mu m)$ are more than 20 times that after the dust storm, while the number concentrations of fine particles $(D < 2 \mu m)$ are only 7 times that after the dust storm.

5. Photochemical reaction mechanism of ozone variations in the surface layer

The change of tropospheric ozone is also a factor affecting the climate. One of the primary sources of tropospheric ozone is from the photochemical reaction of anthropogenic pollutants (McKeen et al., 1991). Most anthropogenic and natural pollutants, such as NO_x , NMHC (non-methane hydrocarbons), CO, etc., are directly emitted to the low troposphere, and are increasing year after year (Berntsen et al., 1995). Most research has focused on urban regions, but little on rural areas. So the mechanism of ozone variation in clean areas are not clear yet. On the basis of meteorological fields provided by a mesoscale nonhydrostatic model (MM5), a three-dimensional regional chemical model (RADM) was applied to China to address the photochemical reaction mechanisms of surface ozone (Yang et al., 1999; Yang and Li, 1998). Below are the main conclusions:

- (1) In polluted areas, surface ozone is primarily dependent on photochemical reactions. This is due to the higher concentration of ozone precursors such as NO_x , NMHC, CO, etc. Physical factors such as transport and diffusion are secondary in determining surface ozone. But in clean areas, the photochemical reaction is not the controlling factor. Surface ozone on the Tibetan Plateau is sensitive to ozone perturbation in upper layers. So it is suggested that the higher surface ozone concentration in summer on the Tibetan Plateau is mainly due to the high background ozone level above the planetary boundary layer which can be carried to surface layer by vertical diffusion and transport.
- (2) OH and HO₂ radicals are the primary atmospheric oxidants and determine the lifetime of most

substances in the atmosphere. They play key roles in the tropospheric photochemistry. At the same time, the variation of ozone precursors such as NO_x , NMHC, and CO, which are largely influenced by human activities, may directly or indirectly affect the concentration of OH and HO_2 . Based on theory and model analysis, we found that the feedback effect of ozone variation on OH and HO_2 radicals is very important (Yang et al., 1999).

(3) There is a complicated nonlinear relationship between surface ozone and NO_x . This nonlinear relationship not only influences the horizontal distribution of surface ozone but also its vertical profile, especially in the heavily polluted areas. Where there is high NO_x pollution, there may be higher ozone concentrations in the upper layer (Yang et al., 1999). During midday in summer, when the steady state is achieved, the ratio of O_3 to $\mathrm{NO}_2/\mathrm{NO}$ has a constant value of 15:1, which can be deduced by the photochemistry equilibrium theory of O_3 , NO_2 , and NO_2 .

6. Long time observation of atmospheric CO_2 and CH_4

During 1985–1988, atmospheric CO₂ and CH₄ in Mingin, Gansu province were continuously observed. The data show that CO_2 concentration experienced an increasing trend of 0.3% per year; CH₄ concentration had evident seasonal variation with an average increasing trend of 1.7% per year (Wang et al., 1989). A project to monitor the concentration of atmospheric CH₄ and its long-term changes has been carried out in Beijing, China since 1985. The data show that atmospheric CH₄ in Beijing is still increasing, although its increasing rate has significantly decreased from the average value of 1.76% per year during 1985–1989 to 0.50% during 1990–1997 (Wang, 1999). The seasonal variation of CH₄ shows a double-peak pattern, with one peak appearing in winter and the other in summer. After 1993, the annual seasonal increasing rate of CH_4 in summer is negative while the increasing rate in winter (due to the emission from non-biogenic sources, such as fossil fuel combustion) is positive and about 25 ppbv per year. As a result, the increase of CH₄ emission from non-biogenic sources in winter is the major cause of the annual seasonal increasing rate from 1993 to 1997 (Wang and Wang, 2000). Concentrations of CO₂, and N₂O have been measured weekly in Beijing from 1993 to 2002. CO₂ concentration in Beijing was about 400 ppm on average from 1993 to 2002, 35 ppm higher than the background concentration (Waliguan, China). Concentrations of CO₂ in Beijing have risen slowly in the past ten years, where the average increasing trend was 0.57% per year; N₂O concentration in Beijing was about 320 ppm on average from 1993 to 2002. Concentrations of N_2O in Beijing were nearly stable from 1993–1998, but they increased rapidly from 1998 to 2002, with an average increasing trend of 2.4% per year.

7. Analysis of trace organic components in the atmosphere

The concentrations of trace organic components in the atmosphere, such as NMHC, CFCs, HCFCs, etc., is very low, but they have important effects on the climate. An automatic system developed by us to monitor trace chlorofluorocarbons (CFCs) and benzene, toluene, xylene, ethylbenzene (BTXE) and NMHC in Xinglong, Mingin, Dinghushan and Beijing, consists of an Ion Trap-GC/MS (gas chromatograph/mass spectrometer) and an intelligent interface for cryoconcentrating and sampling trace organic components of the atmosphere (ICCS). We, for the first time, obtained the concentrations of 36 organic pollutants in the atmosphere baseline in China (Wang and Shi, 1996). The automatic system to monitor NMHC and the other volatile organic polltants of the urban boundary layer of the atmosphere consists of Quadrupole-GC/MS and the ICCS, too. ICCS can concentrate a $500-2000 \text{ cm}^3$ air sample in the analysis process. The instrument provides bulk composition measurements with a detection limit of 1 pptv. In other words, the lowest detection limit of the commercial instrument GUMS is lowered from 1 ppmt to 1 pptv with ICCS. ICCS-GC/MS can monitor all of the organic compounds which have concentrations over 1 pptv. One of the systems is used to monitor the trace volatile organic components of the atmosphere in Beijing. About 25 species of CFCs, 30 species of BTXEs, and 50 species of other NMHCs can be routinely monitored by the automatic system.

8. Emission of HFCs, PFCs, and SF_6 from China in 1995

Based on the investigation of industry production data and consumption data, the emissions of hydrofluorocarbons (HFCs), perfluorocarbon (PFCs), and sulfur hexafluoride (SF₆) from China in 1995 are estimated primarily to be 2244, 2581 and 215 tons, which account for 0.9%, 6.5%, and 3.7% of the global total emissions, respectively (Zhang et al., 2000).

9. Scientific questions and future topics

During the past decade, the distributions of a number of radiatively-active substances has been identified and quantified. However, in most cases, the level of scientific understanding in the calculated radiative forcing of these various substances is still extremely low. In order to reduce the uncertainties in the calculated

radiative forcing of the climate system, the questions listed below need further research:

- (a) What are the relative roles of stratosphere-troposphere exchange, anthropogenic and natural precursor emissions, in situ photochemical processes, and tropospheric transport in controlling ozone and its effect on climate change? Global measurements of ozone and ozone precursors should be carried out. There are several approaches needed for global ozone mapping: satellite, in-situ, and modeling.
- (b) What are the sources, sinks, distributions, and properties of aerosol particles and their direct radiative effects on climate? This seeks the best estimate of existing aerosol optical depth (AOD) and seeks to attribute existing AOD in terms of major aerosol components—sulfates, organic compounds, black carbon, nitrates, dust, and sea salt. Achieving this global characterization will require the integration of aerosol sensing satellites, chemical transport models, and insitu measurements.
- (c) What are the effects of aerosol particles on clouds, their optical properties, precipitation, and regional hydrological cycles? We must have a combination of in situ measurements of cloud and aerosol microphysics and chemistry, both detailed process and large-scale models, and airborne and satellite remote sensing measurements of cloud radiative characteristics to arrive at a better understanding of the processes relating aerosol physical and chemical properties, cloud albedo, and precipitation development.
- (d) How will changing emissions and deposition of gases and aerosol particles affect spatial patterns of climate forcing? Works on this topic will include: more robust validation of climate models, more accurate climate projections for the future, and more comprehensive policy tools for managing emissions.

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