Chinese Science Bulletin

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Springer

Methane emissions from terrestrial plants over China and their effects on methane concentrations in lower troposphere

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Methane (CH_4) is the most important greenhouse gas and reactive trace gas in the atmosphere. Recently, it has been reported that terrestrial plants can emit CH_4 under aerobic conditions, which may call for reevaluation of the inventory of CH_4 emissions in China. In this paper, those emissions over China and their effects on CH_4 concentrations in lower troposphere were investigated. Firstly, based on the work of Keppler et al., the aerobic plant CH_4 emission model (PLANTCH4) for China was established. And by using the database of normalized difference vegetation index (NDVI) derived from NOAA/AVHRR, the distribution of net primary productivity (NPP) over China was simulated, and thereby, for the first time, the amount and distribution of the newly identified source in China were estimated. Secondly, with the aid of the three-dimensional atmospheric chemistry model system (MM5-CALGRID), the effects of the emissions were studied. The results show that the annual aerobic plant CH4 emissions over China amount to 11.83 Tg, i.e. nearly 24% of Chinese total CH_4 emissions. And the major fraction (about 43%) comes from forests. When those emissions are considered in modeling, computed countrywide mean surface concentration of CH_4 is 29.9% higher than without them, with a maximum increase of 69.61 $\mu g \cdot m^{-3}$ in the south of Yunnan Province. In conclusion, to study CH_4 emissions from terrestrial plants over China may have important implications for correctly estimating the contribution of China to global CH_4 budget, and may call for a reconsideration of the role of CH_4 in global and regional environment and climate change.

aerobic conditions, terrestrial plants, methane emissions, normalized difference vegetation index, net primary productivity

Methane (CH_4), with a current globally-averaged mixing ratio of 1.774 ppmv[1], is not only the most abundant organic trace gas in the atmosphere, but also the most abundant reactive trace gas in the troposphere[2]. On the one hand, because the oxidation of CH_4 by hydroxyl radical (OH) deeply affects the concentration of OH, formaldehyde (CH_2O), carbon monoxide (CO) and ozone (O_3) in the troposphere[2], CH_4 plays a significant role in the global and regional photochemical chemistry, especially in nonurban areas[3]. On the other hand, considering that CH_4 is one of the most important greenhouse gases and has 15—30 times greater infrared absorbing capacity than carbon dioxide (CO_2)[4] on a mass basis, it is a key species affecting the energy budget of earth-atmosphere system and climate. Consequently, the investigations on the budget and atmospheric chemistry characteristics of CH_4 are always receiving much concern.

After nearly 20 years of study, it is generally believed that atmospheric CH_4 originates from both non-biogenic and biogenic sources. Biogenic sources include rice paddies, wetlands, termites, livestocks, biomass burning

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Received January 25, 2008; accepted July 30, 2008; published online October 15, 2008
doi: 10.1007/s11434-008-0402-6

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Supported by National High-Tech Research & Development Program of China (Grant No. 2006AA06A307), National Basic Research Program of China (Grant Nos. 2006CB403706 and 2006CB403703), and the Program for New Century Excellent Talents in University, and Nanjing University Talent Development Foundation

etc., while non-biogenic ones include fossil fuel burning, landfills, seepage from fossil fuel mining, extraction, transportation, etc.[5]. Nevertheless, recent findings have demonstrated that our commonly accepted global CH_4 budget is limited. Firstly, Frankenberg et al.[6] compared CH_4 concentration fields retrieved from SCIAMACHY with those simulated with use of a global chemistry transport model, and pointed out that methane sources over tropical rainforests may be considerably underestimated. Secondly, Ferretti et al.[7] reported a surprising variation in the stable carbon isotope values of CH_4 $\delta^{13}C$ ($^{13}C/^{12}C$) in Antarctic ice cores from 1000 to 1500 AD, which was about 3‰ higher than that simulated from currently used preindustrial methane budgets. Thirdly, Carmo et al.[8] studied CH_4 emissions from upland forests in the Brazilian Amazon by observation and modeling, and found an unidentified source of CH_4 in the range of 4—38 Tg/a there. All the discoveries mentioned above indicated that there were still some unknown CH_4 sources in the nature. Thus, Keppler et al.[9] investigated the possibility of methane formation by plant material. After measuring CH_4 release rates and $\delta^{13}C$ values during incubation experiments, they announced that terrestrial plants indeed give off CH_4 under aerobic conditions. And when they extrapolated the fluxes to global vegetation, it seemed that plants could account for 10% to 30% of total CH_4 emissions. Though Dueck et al.[10] argued to the contrary, most researchers were on the side of Keppler et al. Apart from the findings reported by Frankenberg et al., Ferretti et al. and Carmo et al., Crutzen et al.[11] reinterpreted 1988 field data from the Venezuelan savanna region, and strongly supported that plants were the major source of CH_4. Furthermore, they used those data to deduce that the CH_4 emissions from world's tropical savannahs were 30—60 Tg/a, similar to the estimate of Keppler et al.

Heretofore, the physiological process related to CH_4 formation in terrestrial plants under aerobic conditions is unknown. To quantify the newly established CH_4 source, Keppler et al. made the assumption that the global emissions can be scaled relative to four factors, that is the measured emission rate, the global annual net primary production (NPP), the length of vegetation period and the averaged daily sunshine hour. They distinguished the factors between different vegetation categories, and firstly calculated the number to be in the range of 62.8—242.6 Tg/a (average 152.2 Tg/a). Subsequently, more and more researchers engaged in the quantification as well. Parsons et al.[12] re-estimated the amount by using standing leafy biomass and non-leafy biomass instead of biome annual NPP. Their re-calculated value was 52.7 Tg/a. Analogously, Kirschbaum et al.[13] used two modified methods, namely leaf-mass-based method and photosynthesis-based method, to re-estimate the amount as well. Their estimate was in the order of 10—60 Tg/a. In addition, Houweling et al.[14] compared the results from the atmospheric transport model (TM3) with and without plant CH_4 emissions with SCIAMACHY CH_4 retrievals, and derived the upper limit of the source to be 125 Tg/a. Ferretti et al.[15] analyzed the ice core records of atmospheric CH_4 concentration and $\delta^{13}C$ values over the last 2000 years, and determined the emissions to be 0—176 Tg/a. From their results, it seems that aerobic plant CH_4 emissions may have important implications for the global CH_4 budget in despite of the estimation uncertainties.

The above newest research progress in aerobic plant CH_4 emission may have not only some referential meaning to the consummation of the Chinese CH_4 emission inventory, but also certain promotional effect to the study of climate and environment change in China. Unfortunately, the present series of studies tended to focus on the global total amount rather than the distribution, let alone the amount and distribution in China. It would seem, therefore, that further investigations are needed in order to ameliorate the estimate method and hereby to study the emissions over China. The purpose of this paper is to do so. We first present the Chinese aerobic plant CH_4 emission model (PLANTCH4) established based on the work of Keppler et al. Then we estimate the amount and distribution in China by using the detail NPP data calculated from the satellite NDVI data. Finally, we run the three-dimensional atmospheric chemistry model system (MM5-CALGRID) with and without the emissions to quantitatively investigate the effect of them on CH_4 concentrations in lower troposphere in China.

1 Methodology

In this work, the Chinese aerobic plant CH_4 emission model (PLANTCH4) and the atmospheric chemistry model system (MM5-CALGRID) were combined to simulate. The base year 2000 was selected. The model domain extended from 90.6°E to 151.8°E and from 18.6°N to 53.5°N, covering most of China.

1.1 Aerobic plant methane emission model

Based on the method presented by Keppler et al., Chinese aerobic plant CH_4 emission model (PLANTCH4) was built. The model was applied to calculate the emissions at the center of 3696 75km × 75km cells distributed in a horizontal grid of 66 rows and 56 columns. The vegetation category of each cell was decided by the dominant vegetation in it. The detailed data were from 25-category USGS (United States Geological Survey) vegetation data with the resolution of 2 minutes. According to the category, the emission in each cell was estimated by the emission factor, the annual NPP, the length of vegetation period and the averaged daily sunshine hour. Thus, the total emission in China was calculated by using the following equation:

$$P(\mathrm{CH_4})_{\mathrm{annual}} = \sum_{\mathrm{Grid}} \mathrm{NPP} \times (\mathrm{SL} \times \mathrm{ER}_{\mathrm{day\_living}} + \mathrm{Period} \times \mathrm{ER}_{\mathrm{day\_litter}}), \quad (1)$$

where $P(\mathrm{CH_4})_{\mathrm{annual}}$ is the summed-up annual production of CH_4 over the gird (g/a); NPP, SL and Period are the annual NPP (10^{12}g DW/a), plant growing season length (*d*) and period of plant decay (*d*) in a certain cell, respectively; $\mathrm{ER}_{\mathrm{day\_living}}$ and $\mathrm{ER}_{\mathrm{day\_litter}}$ are average daily CH_4 emission rates (10^{-9}g/(g DW·d)) for living plants and detached leaves, and can be calculated as:

$$\mathrm{ER}_{\mathrm{day\_living}} = (\mathrm{ER}_{\mathrm{sun\_living}} \times h_{\mathrm{sun}}) + (\mathrm{ER}_{\mathrm{nosun\_living}} \times (24 - h_{\mathrm{sun}})), \quad (2)$$

$$\mathrm{ER}_{\mathrm{day\_litter}} = (\mathrm{ER}_{\mathrm{sun\_litter}} \times h_{\mathrm{sun}}) + (\mathrm{ER}_{\mathrm{nosun\_litter}} \times (24 - h_{\mathrm{sun}})), \quad (3)$$

where $\mathrm{ER}_{\mathrm{sun\_living}}$ and $\mathrm{ER}_{\mathrm{nosun\_living}}$ are the emission rates of CH_4 from living plants with and without direct sunshine (10^{-9} g/(g DW·d)), respectively; $\mathrm{ER}_{\mathrm{sun\_litter}}$ and $\mathrm{ER}_{\mathrm{nosun\_litter}}$ are the values for leaf litter; h_{sun} is the daily sunshine hour (h/d).

The values of SL, Period, $\mathrm{ER}_{\mathrm{sun\_living}}$, $\mathrm{ER}_{\mathrm{nosun\_living}}$, $\mathrm{ER}_{\mathrm{sun\_litter}}$, $\mathrm{ER}_{\mathrm{nosun\_litter}}$ and h_{sun} of different vegetations used in eqs. (1)—(3) came from the work of Keppler et al. For all plants, mean (low/high) values of $\mathrm{ER}_{\mathrm{sun\_living}}$, $\mathrm{ER}_{\mathrm{nosun\_living}}$, $\mathrm{ER}_{\mathrm{sun\_litter}}$ and $\mathrm{ER}_{\mathrm{nosun\_litter}}$ were 374(198/598), 8.7(1.6/15.8), 119(30.7/207) and 1.6(0.1/4.4) 10^{-9} g/(g DW·d), respectively. Nevertheless, according to different characteristics, each vegetation category was attributed individual SL, Period and h_{sun} values (see Table 1).

The NPP in eq. (1) is the most important factor to distinguish the emissions in different cells. So, it needs to be calculated over the grid to obtain the emission distribution. There are three basic approaches used to model terrestrial NPP, that is statistic-based models, parametric models and process-based models[16]. However, many of them have demerits in application to China. At present, utilizing remote sensing data from meteorological satellite is a good objective way to estimate temporal and spatial distribution of NPP in China, in virtue of the timeliness, dynamic, veracity, extensiveness of satellite data. In previous investigations, Xiao et al.[17] and Zheng et al.[18] established the relationship between the NPP in China and the NDVI derived from NOAA/AVHRR. Based on their results, we estimated the annual NPP in each cell as

$$\mathrm{NPP} = -0.6394 - 67.064 \times \ln(1 - \mathrm{ANDVI}), \quad (4)$$

where ANDVI is the annual average value of NDVI. NDVI is the dimensionless index calculated from the satellite data in the red and near-infrared, which is related to vegetation coverage, growth and photosynthetic capacity. The NDVI data used here were from USA NASA Pathfinder AVHRR, covering the area 70.0°E to 140.08°E and 5.0°N to 55.0°N with the resolution of 0.072°. There were three steps to calculate ANDVI. Firstly, maximum value of NDVI in every ten days was chosen to eliminate cloud effects[18]. Secondly, three-point smoothing filtering was used to avoid other noises[18]. Finally, the ANDVI was calculated as

$$\mathrm{ANDVI} = \frac{1}{36} \sum_{i=1}^{36} \mathrm{NDVI}(i). \quad (5)$$

Table 1 Vegetation categories, and their SL, period and sunshine hour

Vegetation category	Season length (d)	Period (d)	Sunshine hour (h/d)
Urban	0	0	0
Arable land (5 types)	200	90	8
Grassland (4 types)	150	90	6
Shrub land	200	180	8
Savanna	200	365	8
Woods (7 types)	250	90	6
Water body	0	0	0
Sparse vegetation	100	90	6
Tundra (2 types)	100	90	6
Snow or ice	0	0	0

1.2 Atmospheric chemistry model system

The atmospheric chemistry model system used in this paper mainly consists of two models, MM5 and CALGRID, with the same grid system used in

PLANTCH4. MM5 was used for meteorology modeling. The pressure at the model top was 100 hPa, and 10 unequispaced levels in the vertical were chosen. Blackadar's high resolution boundary layer scheme, time-dependent boundary condition and Anthes-Kuo deep cumulus convection parameterization were selected in the simulation. CALGRID was used for atmospheric chemistry modeling. The height at the model top was 5000 m, and another 10 unequispaced levels in the vertical were chosen. The gas phase chemical mechanism named SAPRC-90 and the chemical integration solver based on the quasi-steady-state method (QSSA) were used in the current version. The model system took the emissions of NO_x, CO, CH_4, SO_2 and VOCs (10 subcategories) into account. The anthropogenic emission inventory was from Streets et al.'s work[19]. CH_4 emission data from rice paddies were based on the work of Xie et al.[20], and those from aerobic plants were supplied by PLANTCH4. We computed CH_4 concentrations in lower troposphere over China for 15 d per month in January, April, July and September, which represented winter, spring, summer and autumn, respectively. Averaging the results of those days, we got the annual status.

2 Results and discussions

2.1 Net primary productivity in China

Based on the method mentioned in section 1.1, it was estimated that total annual NPP of terrestrial vegetation over China in 2000 is 12.67×10^9t DW/a. Compared with the work of Sun et al.[21], where the value was 7.438×10^9 tDW/a by using Miami model or 5.29×10^9 tDW/a by using light utility efficiency model, our results are a little higher. Considering that the experiential relationship between NDVI and NPP used in this paper was derived from limited observation data, the discrepancy is reasonable and acceptable.

Figure 1(a) illustrates the distribution of annual NPP over China. It shows that the intensities vary significantly among regions. Generally speaking, in the highly forested subregions in China, the NPP values are above 20 tDW/($hm^2\cdot a$). In most agricultural areas, the values are around 10—15 tDW/($hm^2\cdot a$). In part areas of Inner Mongolia, Qinghai, and Tibet, where the dominant vegetation is grassland, the values are about 5 tDW/($hm^2\cdot a$). And in the west of Inner Mongolia and Gansu, the northwest of Qinghai, the south of Xinjiang, where barren land is dominating, the value is very low. Furthermore, by comparing Figure 1(b) with Figure 1(a), we can see that the patterns are similar. This means that our results of NPP coincide with the real situation of ecological system in China.

2.2 Aerobic plant methane emissions over China

With the aid of PLANTCH4, aerobic plant CH_4 emissions over China in 2000 are roughly estimated to be 11.83 Tg/a (in the range of 4.83—19.70 Tg/a), which is about 7.8% of the global total according to the value 152.2 Tg/a reported by Keppler et al.[9]. Streets et al.[19] and Xie et al.[20] studied CH_4 released from anthropogenic sources and rice paddies in China, respectively. According to their results (28.57 Tg/a reported by Streets et al. and 9.26 Tg/a reported by Xie et al.), aerobic plant CH_4 emissions are about 24% of the total CH_4

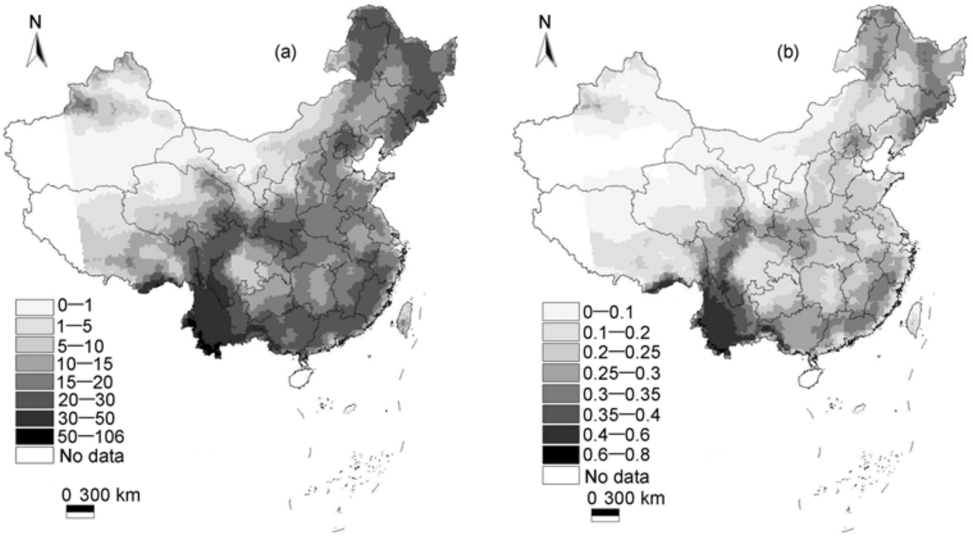

Figure 1 Distribution of annual NPP (a) and ANDVI (b) over China in 2000 (tDW/($hm^2\cdot a$)).

emissions in China, which is closer to the ratio reported by Keppler et al. In addition, it is shown in Table 2 that emission percentages in forests, arable lands, grasslands and others are 43.0%, 28.3%, 19.0% and 9.7%, respectively. Though they are different from the study of Keppler et al., where the percentages are 66.0%, 4.8%, 24.7% and 4.5%, the results of this study should reflect the real distribution of land resources in China on account of the relative abundant of arable lands, the relative scarcity of forest resources and the destruction of grasslands there.

Table 2 Annual aerobic plant methane emissions from major vegetation categories in China (Tg/a)

Vegetation category	Annual CH_4 emission (min/max)
Forests	5.09 (2.04/8.49)
Arable lands	3.35 (1.41/5.56)
Grasslands	2.25 (0.90/3.75)
Others	1.15 (0.48/1.90)
Total	11.83 (4.83/19.70)

Figure 2(a) gives the distribution in China. It shows that the fluxes generally decline from southeast to northwest, and remarkably vary among vegetations. Southwest of Yunnan and south of Tibet are covered with the highest CH_4 emission. And the fluxes there, which are estimated for the total flux from each 75 km × 75 km grid cell, are over 1000 g/s. In the southwest and northeast of Guangxi, the fluxes are around 700—800 g/s. In the south of Shaanxi, the borders between Guangdong and Guangxi, the borders between Guangdong, Jiangxi and Fujian, and the borders between Jiangxi, Fujian and Zhejiang, the fluxes are greater than 500 g/s. It is noteworthy that the dominant vegetation category of the aforementioned areas is forest. In addition, in most areas to the east of 100°E, such as the Northeast Plain, the North China Plain, the middle and lower Yangtze River Plain, the Sichuan Basin, South China, etc., where arable land density is very high, we can see that the fluxes of aerobic plant CH_4 emissions are about 200—400 g/s. And for Northwest China, the fluxes are lower than 100 g/s because of the lack of vegetation coverage.

From the statistical data for every province, Yunnan has the largest contribution in China (13.9%), with the annual total emission amount of 1.65 Tg/a. And that from three northeast provinces (Heilongjiang, Jilin and Liaoning) is 1.73 Tg/a, about 14.6% of the countrywide total. Besides the above provinces, there are considerable emissions in Inner Mongolia, Sichuan, Tibet, Guangxi, etc. as well.

Figure 2(b) shows the distribution of CH_4 emissions from human activities and rice paddies. We can see that there are high fluxes in the Beijing-Tianjin-Tangshan area, Yangtze River Delta region and Pearl River Delta region, where the density of population is big, and the industrial and agricultural sectors are highly developed. It can be deduced that the dominant CH_4 sources in those areas are anthropogenic origin. Moreover, there are also relative high CH_4 emissions in Sichuan, Chongqing, Central China, etc., where the dominant vegetation is arable land. We can deduce that CH_4 in

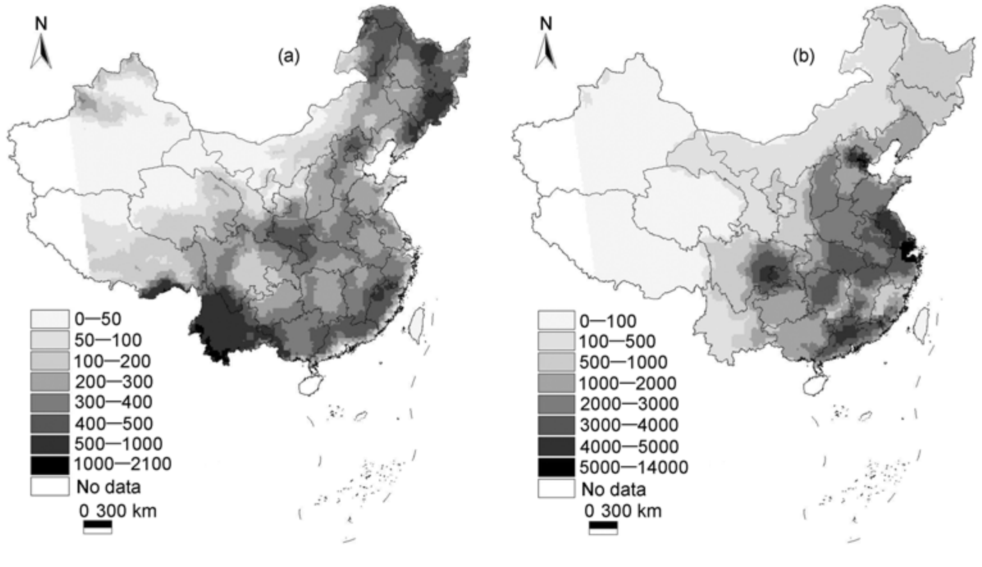

Figure 2 Distribution of methane emissions from aerobic plants (a) and other sources (b) over China in 2000 (g/s).

those areas is mainly released from rice paddies. Additionally, by comparing Figure 2(a) with Figure 2(b), it can be found that aerobic plant emission is the major CH_4 source in the remote and undeveloped areas of China, especially forests.

2.3 Effects on methane concentrations in lower troposphere over China

2.3.1 Modeling schemes. In order to investigate the effects of aerobic plant CH_4 emissions on CH_4 concentrations in lower troposphere over China, two modeling schemes were designed: (1) We only considered CH_4 emissions from anthropogenic sources and rice paddies and (2) we considered CH_4 emissions from all sources, including aerobic plants. The results from scheme (2) are closer to truth. The differences between the CH_4 concentration fields computed from schemes (2) and (1) show the effects expected. In those schemes, the initial and boundary conditions of CH_4 were assigned relatively small value, which means that only contribution of Chinese local sources was considered. To take the contribution of large scale regional sources into account, 1150 μg/m^3 (about 1.6 ppm), in the name of background CH_4 concentration, was added to the results computed from the above schemes.

2.3.2 Effects on CH_4 concentrations near surface. The distribution of annual average CH_4 concentrations over China estimated from scheme (2) is shown in Figure 3(a). It indicates that, owing to the long lifetime of CH_4, the pattern is obviously affected by the processes of atmospheric transportation and diffusion. By comparing Figure 3(a) with Figure 2, however, we still can tell the effects of different sources on CH_4 concentrations. Firstly, those high concentrations (>1300 μg/m^3) in the North China Plain and Shanghai should be mainly caused by human activities. Secondly, the similar values in the Sichuan Basin should be mostly from rice paddies. Thirdly, the relatively high CH_4 concentrations (>1250 μg/m^3) in some areas of Yunnan and Guangxi are markedly from the forests there. Finally, the CH_4 concentrations (>1225 μg/m^3) in the major farming areas in China, such as the Northeast Plain, the middle and lower Yangtze River Plain, South China, etc., are obviously affected by rice paddies.

Figure 3(b) shows the effects of aerobic plant CH_4 emissions on CH_4 concentrations near surface over China. It indicates that the latter are considerably higher when the former are taken into account, and remarkable changes of CH_4 concentration usually take place near the major sources. For example, in Northeast China, the concentration of CH_4 increases more than 20 μg/m^3 as a result of vegetation emission. And due to the contribution of forests, the variation in the south of Tibet and southwest of Yunnan is greater than 50 μg/m^3, with a maximum of 69.68 μg/m^3 in the south of Yunnan, where the fluxes of aerobic plant CH_4 emission are very high. In addition, it is noteworthy that because the distribution of the long lifetime species is deeply affected by atmospheric transportation, the CH_4 concentrations in a large area around the woods observably increase owing to aerobic plant emissions. On the whole, the newly identified source causes 29.9% ((scheme (2)-scheme (1))/scheme (1)) increase of the countrywide mean con-

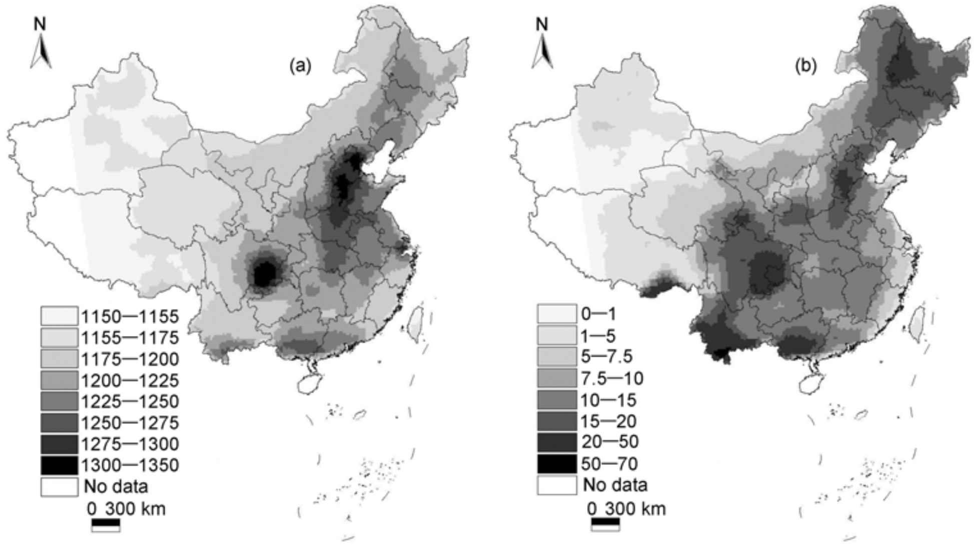

Figure 3 Distribution of annual average concentration of CH_4 near surface over China (a) and the effects of aerobic plant emissions on them (b) (μg/m^3).

centration of CH_4 in China.

3 Conclusions

In this paper, aerobic plant methane emission model (PLANTCH4) for China was established on the basis of the newest advance in the study of CH_4 sources. And the NDVI data derived from NOAA/AVHRR were used to obtain the distribution of NPP in China. Thus, the annual total amount and distribution of CH_4 emissions from terrestrial plants over China were estimated for the first time. Afterwards, with the aid of the three-dimensional atmospheric chemistry model system (MM5-CALGRID), the effects of the above-mentioned emissions on CH_4 concentrations in lower troposphere were investigated. The main conclusions are listed as follows:

(1) The total annual NPP of terrestrial plants over China is 12.67×10^9 tDW/a. The characteristic of distribution is the general decline from southeast to northwest. Forests have the highest NPP value in unit area, followed by arable lands, grasslands and barren areas. Our results of NPP are closer to the real situation of ecological system in China.

(2) The annual CH_4 emissions from terrestrial plants over China amount to 11.83 Tg, with 43.0% from forests, 28.3% from arable lands, 19.0% from grasslands and 9.7% from others. The emissions are about 7.8% of the global total, and account for 24% of total CH_4 emissions in China. A majority of the emissions is from Yunnan Province, with the percentage of 13.9%. In addition, three northeast provinces contribute about 14.6% of the Chinese total. And there are considerable emissions in Inner Mongolia, Sichuan, Tibet, Guangxi etc. as well.

(3) It is obvious that the effects of aerobic plant emissions on CH_4 concentrations in lower troposphere over China are significant. The countrywide mean surface concentration of CH_4 in China increases by 29.9%, with a maximum of 69.68 $\mu g/m^3$ in the south of Yunnan Province.

With the investigations to the depth, the newly identified source may exert an important influence on the currently accepted CH_4 budget, and then may call for a reconsideration of the role of atmospheric CH_4 in global and regional environment and climate change. Unfortunately, because of our limited knowledge at present, the estimation method is not perfect, and large uncertainties in the inventory remain. To overcome those insufficiencies, more additional studies are required. Consequently, further investigations in laboratory and field should be carried out to identify the physiological process, and hereby to consummate a better emission model.

The authors thank Liu Li from Guizhou Institute of Meteorological Sciences for providing the NDVI data derived from NOAA/AVHRR.

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