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Nitrous oxide in the Changjiang (Yangtze River) Estuary and its adjacent marine area: riverine input, sediment release and atmospheric fluxes

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Abstract

Dissolved nitrous oxide (N_2O) was measured in the waters of the Changjiang (Yangtze River) Estuary and its adjacent marine area during five surveys covering the period of 2002–2006. Dissolved N_2O concentrations ranged from 6.04 to 21.3 nM, and indicate seasonal variations with high values occurring in summer and spring. Dissolved riverine N_2O was observed monthly at station Xuliujing of the Changjiang, and ranged from 12.4 to 33.3 nM with an average of 20.8±7.8 nM. The average annual input of N_2O from the Changjiang to the estuary and its adjacent area was estimated to be 15.8×10^6 mol/yr. N_2O emission rates from the sediments of the Changjiang Estuary in spring ranged from −1.88 to 2.02 $\mu mol\,m^{-2}\,d^{-1}$, which suggest that sediment can act as either a source or a sink of N_2O in the Changjiang Estuary. The annual sea to air N_2O fluxes from the Changjiang Estuary were estimated to be 6.8±3.7, 13.3±7.2 and 14.9±8.3 $\mu mol\,m^{-2}\,d^{-1}$ using LM86, W92 and RC01 relationships, respectively. The annual sea to air N_2O fluxes from the adjacent marine area were estimated to be 8.5±7.8, 15.3±13.5 and 17.4±15.7 $\mu mol\,m^{-2}\,d^{-1}$ using LM86, W92 and RC01 relationship, respectively. Hence the Changjiang Estuary and its adjacent marine area is a net source of atmospheric N_2O.

1 Introduction

N_2O is an important trace gas in the atmosphere, which is responsible for 5–6% of the greenhouse effect (Houghton et al., 1996), and also contributes to the destruction of the ozone layer (Crutzen and Schmailzl, 1983). The global atmospheric N_2O concentration has increased from a pre-industrial value of about 270 to 319 ppb in 2005 (IPCC, 2007). The oceans are considered to be significant sources for atmospheric N_2O, contributing to about 25% of the global emissions (Nevison et al., 1995; Bouwman et al., 1995). However, emission of N_2O from the oceans is not uniformly distributed geographically. Estuaries have been subject to intense anthropogenic inputs of inorganic

BGD
7, 3125–3151, 2010

nitrogen as a consequence of fertilizer usage and sewage input over recent decades (Howarth et al., 1996). One important consequence of the increased N load is the enhanced production and emission of N_2O from estuaries (Barnes and Owens, 1998; De Wilde and de Bie, 2000; Marty et al., 2001; LaMontagne et al., 2003; Ganier et al., 2006). Although estuaries represent only about 0.4% of the global ocean area, Bange et al. (1996) estimated they account for about 33% of the oceanic N_2O emission. However, these estimates are rather uncertain due to high spatial and temporal variability and the limited data available, especially for the typical large river estuaries in the world. For example, very limited data are available for the Amazon, the Mississippi and large river estuaries in Asia.

The Changjiang (Yangtze River), 6300 km at total length, is the largest river in Asia, ranking third among the world's rivers. Its drainage basin covers about 1.8 million km^2 in area, which is about one fifth of the total land area of the whole country. The huge amount of runoff discharge (903 km^3/yr averaged from the 1950s to 2005), and sediment load (414 million tons/yr averaged from the 1950s to 2005) enter its estuary and are emptied into the East China Sea (ECS) (Wang et al., 2008). The Changjiang estuary is of particular interest to nutrient cycling because of its importance for the transportation of terrigenous nutrients to the coastal seas. The annual dissolved inorganic nitrogen and phosphate concentrations and fluxes from the Changjiang show a stable to slow increasing trend from the 1950s to the early 1980s, but then increased abruptly (Liu et al., 2003, 2007). For example, the nitrate concentration near the mouth of the Changjiang has increased from ~60 µmol/L in the 1980's to ~80 µmol/L in 1997 (Liu et al., 2003). On a global scale, half of the total nitrogen load received by estuaries has been estimated to be removed by denitrification (Nixon et al., 1996). The Changjiang Estuary also exhibits turbidity maximum at the mouth (Li and Chen, 1998), which is likely to have a significant impact on nitrification in the turbidity maximum (Law et al., 1992). Considering both nitrification and denitrification can produce N_2O, the increasing load of inorganic nitrogen to the Changjiang Estuary may potentially affect the cycling of N_2O, its subsequent emission to the atmosphere and to the open sea as

BGD
7, 3125–3151, 2010

well. However, no data are so far available for the distribution, fluxes and producing processes of N_2O in the seawaters of Changjiang Estuary. Hou et al. (2007) studied the N_2O productions in the intertidal sediments of the Changjiang Estuary, and found that N_2O was mainly from nitrification under the desiccated condition and from denitrification under the waterlogged and reflooded conditions.

We present here a study on the distribution of N_2O in the Changjiang Estuary and its adjacent marine areas. The objectives of study were: (1) to determine the distribution of N_2O along the Changjiang Estuary; (2) to estimate the N_2O emission into the atmosphere; (3) to evaluate the contribution of the Changjiang to the N_2O in the ECS.

2 Methods and materials

2.1 Sample collections

Five cruises were conducted on the Changjiang Estuary and its adjacent area during 25 April–15 May 2002 by R/V "Haijian 47", 4–14 November 2002 by R/V "Science No. 1", during 2–11 June, 19–31 August and 3–13 October 2006 by R/V "Beidou", respectively. The sampling locations are shown in Fig. 1.

Water samples were collected using 10 L Niskin bottles. Subsamples for N_2O determination were transferred from Niskin bottles into 135 mL or 60 mL glass vials using the rubber-connecting tube with a glass pipette end. After overflow of approximately 1.5 to 2 fold of bottle volume, saturated solution of $HgCl_2$ was added to inhibit microbial activity, then the sample vial was immediately sealed with a butyl rubber stopper and an aluminum cap to exclude the excessive water and stored in the dark box. All the water samples were analyzed after returning to the shore laboratory within 60 days of collection. Data of temperature, salinity and dissolved oxygen were obtained from the shipboard CTD profiles.

To quantify the flux of N_2O input to the sea from the Changjiang, N_2O concentrations were monitored monthly at Xuliujing (121°2′ E, 31°46′ N, Fig. 1b), the most downstream

BGD
7, 3125–3151, 2010

main channel station, from May 2007 to April 2008. Since it is located at the further upstream side from limit of salt water intrusion during dry seasons and at the node where the river estuary begins to become wide, Xuliujing is suitable for observing the freshwater input to the sea from the Changjiang. Water samples were collected using 10 L bucket. Subsamples for N_2O determination and the treatment of water samples were the same as above. All the water samples were analyzed within 2 month after collection.

Sediment cores were sampled using a multiple corer (each sediment core 60 cm long and 10 cm I.D.) from stations DC10 and DB6 (Fig. 1a) in the Changjiang Estuary during the spring cruise in 2002. After collection, the cores with 20–30 cm high of sediments were selected and left undisturbed in the plexiglass tubes with the end sealed with air-tight rubber bungs in dark before the determination of trace gas release.

2.2 Chemical analysis

Dissolved N_2O was measured by gas chromatography using a gas-striping method (Zhang et al., 2006). Calibration of the Electron Capture Detector (ECD) responses were done by injection of certain volumes of standard gas of 5.60 ppmv N_2O/N_2 (Research Institute of China National Standard Materials) into the stripper filled with blank seawater. The N_2O of the blank seawater had previously been stripped out together with other dissolved gases by ultra-pure N_2. After injection, the blank seawater was subsequently analyzed by the same procedure used for unknown samples. The Method Detection Limit (MDL) for N_2O analysis in this study was 0.1 nmol/L (MDL is defined as the N_2O concentration in 135 mL seawater sample corresponding to two standard deviations of seven replicates of the blank), respectively. The precisions of repeated analysis of water samples were about 5% for N_2O in routine sample analysis.

Water samples for determination of nutrients were filtered through acid-cleaned acetate cellulose filters (pore size: 0.45 µm). The filtrates were poisoned by $HgCl_2$ and stored in the dark at 4 °C. In the laboratory, nutrients were determined photometrically by an auto-analyzer (Model: Skalar SAN^{plus}) with precision of <5–10%.

BGD
7, 3125–3151, 2010

2.3 Measurements of N_2O emissions from the sediments

N_2O emissions from the sediments of the Changjiang Estuary were determined using closed chamber technique (Barnes and Owens, 1998) during April/May 2002. Measurements were conducted immediately after core collection. After removing the overlying waters carefully without disturbing the biological activity, filtered bottom waters were added carefully without gas phase left. The top of the tubes was then sealed with air-tight rubber bungs equipped with two stopcocks. An aerated pump was put in half of the water phase to stir the water phase. During 2 days incubation experiments, overlying water samples were carefully taken out at 4 h interval through one stopcock fitted in the rubber bang for determination of dissolved O_2, and N_2O changes in the enclosed water phase. The water overlying the core was replenished simultaneously via another stopcock fitted with a syringe containing filtered bottom water. No air was involved during these processes. After each sampling, two bottles of filtered bottom water using as replenishment were collected for determination of the N_2O concentration to correct the N_2O change of the overlying water. The dissolved N_2O in the overlying water were analyzed using the gas-stripping method described above. The dissolved O_2 was measured by a DO probe Model 9101Y (Jenco, USA). The emission rates of N_2O from the sediments were determined from the slope of the N_2O increase in the overlying water versus incubation time.

2.4 Computation of sea-to-air fluxes

Sea to air N_2O fluxes (F in mol m^{-2} d^{-1}) can be estimated by the following equation

$$F = k_w \times (C_{obs} - C_{eq}) \quad (1)$$

Where C_{obs} is the observed concentration of dissolved N_2O; C_{eq} is the air-equilibrated seawater N_2O concentration, which was calculated for in-situ temperatures and salinities using the solubility data of Weiss and Price (1980). Atmospheric N_2O was not measured during these cruises, and a global mean atmospheric N_2O mixing ratio of

BGD
7, 3125–3151, 2010

317.6 ppb for 2002 and 320.3 ppb for 2006 from the NOAA/ESRL halocarbons in situ program (http://www.esrl.noaa.gov/gmd) was used for the calculations in this study. k_w is gas transfer velocity, which is usually expressed as a function of the wind speed (Raymond and Cole, 2001). Various empirical relationships have been derived for estimating k_w. The two most widely used are those of Liss and Merlivat (1986) and Wanninkhof (1992), which are often assumed to define the lower and upper limits for k_w. However, tidal currents may also contribute to water turbulence, especially in inner estuaries with shallow waters and high frictions on the bottom (Raymond and Cole, 2001; Zappa et al., 2003; Abril and Borges, 2004). Raymond and Cole (2001) derived a relationship (k_{600}=1.91 exp(0.35 u_{10})) based on a compilation of published k_{600} values in various rivers and estuaries and obtained using different methods (floating chamber, natural tracers – CFC, ^{222}Rn – and purposeful tracer – SF_6). Their studies suggested that k could be significantly higher in estuaries than in open oceanic waters at the same wind speed. Since no direct measurements of gas transfer velocity were made in the Changjiang Estuary, the relationships of Liss and Merlivat (1986) (hereafter referred to as LM86), Wanninkhof (1992) (hereafter referred to as W92) and Raymond and Cole (2001) (hereafter referred to as RC01) were used to compute k_w to help comparisons with published data. The transfer coefficient was adjusted by multiplying with $(Sc/600)^{-n}$ for LM86 (n=1/2 for wind speed >3.6 m/s and n=2/3 for wind speed <3.6 m/s), $(Sc/660)^{-1/2}$ for W92, and $(Sc/600)^{-1/2}$ for RC01. Sc was calculated according to the equation by Wanninkhof (1992).

The major uncertainty in the assessment of sea to air gas fluxes is related to the estimation of the gas transfer coefficient, which depends on the type of wind data used. In this work, we computed the gas transfer coefficients using averaged monthly wind speeds obtained from the monitoring results beyond the Changjiang Estuary in 1977–1986, which was 6.7 m/s for May, 6.8 m/s for June, 7.6 m/s for August, 7.2 m/s for October and 7.6 m/s for November (cf. Xu, 1992). Since during the May 2002 and October 2006 cruises, wind speeds were continuously recorded shipboard using an automated weather station (Campbell Scientifics, UK), the gas transfer coefficients

BGD
7, 3125–3151, 2010

and sea to air fluxes were also estimated using ship based in situ wind speeds to help comparisons.

3 Results and discussion

3.1 Distributions of N_2O in the Changjiang Estuary and its adjacent area

N_2O concentrations and saturations in the surface and bottom waters of the Changjiang Estuary and its adjacent area during the five cruises from 2002 to 2006 are shown in Table 1, which indicates apparently seasonal variations with the high values occurring in summer and spring. Table 2 shows previously published data on N_2O in other estuaries, which indicates that N_2O concentrations vary over a wide range of 2–1457 nM at various temporal and spatial scales. N_2O in the Changjiang Estuary and its adjacent area falls within this range but toward the low end. Concentrations and emissions of N_2O in estuaries are generally found to be related to the estuarine Dissolved Inorganic Nitrogen (DIN) levels (Seitzinger and Kroeze, 1998; Dong et al., 2004). Mean DIN concentrations in the Changjiang estuary in this study were 12.5±5.8, 12.6±8.6 and 15.6±8.0 µM for June, August and October 2006, respectively. Hence the low N_2O observed in this study is consistent with the low DIN levels (Fig. 2). Another reason for the observed low N_2O in this study may be partly due to that most previously published N_2O data in Table 2 were obtained from inner estuaries while N_2O data in this study were from the outer estuary of the Changjiang. High N_2O were usually observed at the inner estuaries, especially at the low salinity in the vicinity of Turbidity Maximum Zone (TMZ) (Barnes and Owens, 1999; Law et al., 1992; Abril et al., 2000). Turbidity maximum existed all year round in the river mouth of the Changjiang (Li and Chen, 1998). However, given that the sampled salinity range was mainly limited to high salinity of 20–34 and the TMZ was not covered during all surveys, observed N_2O concentrations in the Changjiang estuary in this study only represent the low limit. For example, high concentrations of N_2O, ranging from 26.04 to 37.20 nM with an average

BGD
7, 3125–3151, 2010

of 30.58±4.17 nM, were found at the low salinity area (s of 0.6–13.5) near the mouth of the Changjiang estuary in September 2003 (Zhang et al., 2008). In any way, this study highlights the importance of studies on the N_2O variability of more typical estuaries in the future research to estimate accurately the contribution of estuaries to atmospheric N_2O.

The horizontal distributions of N_2O concentrations in the Changjiang Estuary and its adjacent area are shown in Fig. 3, which indicates great temperal variation. But generally high N_2O concentrations were observed beyond the mouth of the Changjiang and Hangzhou Bay, especially in the bottom waters. N_2O concentrations showed no correlation with salinity. Similar phenomenon has been observed by Amouroux et al. (2002) in the surface water of the north-west Black sea shelf and by Bange et al. (1998) in the estuarine and coastal waters of southern Baltic Sea.

3.2 Riverine input of N_2O

Figure 4 shows the seasonal variation of dissolved N_2O observed at Station Xuliujing during the period of 2007–2008, which ranged from 12.4 to 33.3 nM with an average of 20.6±7.5 nM. Since no regular monitor of flow rates were made at Station Xuliujing, flow rates at Station Datong (about 600 km upstream from Xuliujing) were usually used to represent the water discharge to the sea from the Changjiang. N_2O concentrations in the river waters showed obvious seasonal variations with higher values occurring in both the summer and winter. N_2O concentrations had no correlation with the flow rate but correlated negatively with the in situ temperature ($[N_2O]=-0.5\,t+27.2$, $r^2=0.44$, $n=10$). Table 3 compiled previously published N_2O concentrations and saturations in various world rivers. It can be seen that N_2O concentrations in river waters show great spatial and temporal variations with the range from 3.3 to 527 nM. N_2O concentrations in the surface waters of the Changjiang in this study fall within the reported N_2O ranges in the worldwide rivers. N_2O in the river waters may come from in situ production by nitrification or denitrification (Yan et al., 2004), production and emission by the sediment (García-Ruiz et al., 1999), runoff and ground water from agricultural soils (McMahon

BGD
7, 3125–3151, 2010

and Dennehy, 1999). For example, Yan et al. (2004) reported a N_2O production rate of 1.30±0.85 and 346±261 nmol N m^{-2} h^{-1} by denitrification in the Changjiang for August and October 2002, respectively. The observed N_2O concentration of 19.5 nM in August 2007 in this study was higher than the value of 9.02±1.03 nM observed at station Datong in August 2002 reported by Yan et al. (2004). The observed N_2O concentration of 13.6 nM in October 2007 in this study was in agreement with the value of 13.39±8.71 nM at station Datong in October 2002 reported by Yan et al. (2004). A survey to the mainstream of Changjiang in January 2008 showed that N_2O concentrations from station Yichang to Xuliujing ranged from 16.6 to 30.7 nM with an average of 22.0±3.5 nM (Zhao et al., 2009).

We estimate the annual average input of N_2O from the Changjiang to the estuary and its adjacent area by multiplying the monthly river water N_2O concentration by the monthly flow rate, which yields a N_2O flux of 0.50 mol/s equal to 15.8×10^6 mol/yr for the annual input, and indicates that the Changjiang is an important source for N_2O in the estuary and its adjacent area.

3.3 Sediment release of N_2O

In estuarine and coastal regions, the sediments are likely to be important sources of N_2O emitted to the water column since they are active sites for both nitrification and denitrification (Capone, 1991; Usui et al., 2001). A study by Capone suggests the fluxes from the sediments (predominantly in coastal areas) could account for over 40% of the net oceanic N_2O production (Capone, 1991). High nutrient loading from terrestrial environments together with a close benthic and pelagic coupling due to shallow water depth stimulates microbial processes including N_2O production (Seitzinger and Nixon, 1985; Bange et al., 1996; Seitzinger and Kroeze, 1998). Since the Changjiang Estuary contains large amount of inorganic nitrogen species, one would expect high production and release of N_2O from the sediments.

BGD
7, 3125–3151, 2010

Incubation experiments at stations DC10 (Clay, depth 10 m) and DB6 (Silt, depth 23 m) in May 2002 showed different results. Obvious accumulation of N_2O together with the decrease of O_2 in the overlying water was observed during sediment incubation at station DB6. Sediment oxygen consumption rates and benthic N_2O fluxes from the sediments were calculated to be 32.9 mmol m^{-2} d^{-1} and 2.02 µmol m^{-2} d^{-1}, respectively. Incubation experiments of nutrient exchange between sediment-water interface under oxic environments showed that nitrate was transferred from the sediment to water column, while NH_4^+ was transferred from the water column to sediment at station DB6 (Qi et al., 2003). Since the dissolved oxygen in bottom water of station DB6 is highly saturated (104%, 9.54 mg/L) and the sediment type is silt, easily availabilities of O_2 and NH_4^+ provide favorable conditions for the occurrence of nitrification in the sediments. Hence N_2O may be produced mainly via nitrification in the surface sediments at station DB6 and then released to the water column together with the produced nitrate. During incubation at station DC10 both N_2O and O_2 in the overlying water decreased. Sediment oxygen consumption rates and benthic N_2O fluxes from the sediments were calculated to be 66.8 mmol m^{-2} d^{-1} and −1.88 µmol m^{-2} d^{-1}, respectively, for station DC10. Incubation experiments of nutrient exchange between sediment-water interface under oxic environments showed that both nitrate and NH_4^+ was transferred from the water column to sediment. Since the bottom water of station DC10 is characterized of low dissolved oxygen (4.41 mg/L) and the sediment is clay, O_2 is not easily transferred to the sediment. These suggest that N_2O can be produced via both nitrification and denitrification in the sediment at station DC10, but the occurrence of N_2O reduction simultaneously with the production by denitrification induced net consumption of N_2O in the sediment. Li et al. (2009) reported that the sediment denitrification rates ranged from 101.3 to 731.9 µmol N_2O m^{-2} h^{-1} in June 2006 in the Changjiang Estuary.

Benthic N_2O fluxes obtained in this study fell within the range reported for eutrophic nearshore sediments (−4.8 to 134.9 µ mol N_2O m^{-2} d^{-1}: Usui et al., 1998), and lower than those reported by Wang et al. (2007) for the intertidal sediments of the Changjiang Estuary in summer (1.2–102 µmol N_2O m^{-2} d^{-1}). High N_2O emission rates from the

BGD
7, 3125–3151, 2010

interdidal sediments may be resulted from the increase supplies of nutrients due to bioturbation and bio-irrigation (Barnes and Owens, 1998). Usui et al. (1998) calculated the N_2O fluxes from the sediments to range from -0.516 to $-0.156\,\mu mol\,N_2O\,m^{-2}\,d^{-1}$ at the continental shelf and from -0.264 to $0.444\,\mu mol\,N_2O\,m^{-2}\,d^{-1}$ at the slope region of the East China Sea, the magnitude is lower than benthic fluxes of N_2O in the Changjiang Estuary and suggests that the Changjiang Estuary can act as a stronger N_2O source or sink than the East China Sea. In any way, this study suggests that sediment can act as either a source or a sink of N_2O in the water column in the Changjiang Estuary.

3.4 Air-sea fluxes of N_2O

N_2O saturations in the surface waters of the Changjiang Estuary and its adjacent area ranged from 84% to 363% during the five surveys in this study (Table 1), which showed that the surface waters of studied regions were generally supersaturated with respect to the atmospheric N_2O concentrations all year except a few stations during November 2002. Hence the Changjiang Estuaries and its adjacent areas represent a net source of N_2O to the atmosphere all year around.

The studied area was divided into two areas (i.e. estuary and marine area) according to the salinity of 30. For each station we calculated sea to air N_2O flux based on the actual saturation value and the long term averaged wind speed and the results are summarized in Table 4. From Table 4, it can be seen that the greatest uncertainty for the sea-to-air N_2O flux estimation came from the estimation of gas exchange coefficient. Using different relationships yield significantly different transfer coefficients under the same wind speed. Generally using LM86 relationship yields a lower value, and using W92 and RC01 relationships lead to relatively higher N_2O flux estimates than using LM86 by a mean factor of about 1.9 and 2.2, respectively. Another important uncertainty in the assessment of the gas transfer coefficient and sea-air gas fluxes is related to the type of wind data used. Morell et al. (2001) found that fluxes computed using climatological wind speed data often exceed those using ship-based wind speed

BGD
7, 3125–3151, 2010

measurements by over 50%. In this work, we computed the gas transfer coefficients for May 2002 and October 2006 using both ship-based in situ wind speed and long term averaged wind speed. The long term averaged wind speeds in the studied regions were higher than the mean ship-based in situ wind speeds by 10–130%, hence the obtained N_2O fluxes estimated using long term wind speed were higher than those using in situ wind speed by 20–280%. The discrepancy between all these sets of flux values suggests that we should pay much attention to the uncertainty involved by using different sea-air exchange models and wind speed data when making comparisons with published data. In this paper only sea to air N_2O fluxes estimated using long term wind speed will be discussed below, hence our results should be relatively overestimated to some extent.

The sea to air N_2O fluxes from the Changjiang Estuary and its adjacent marine area indicated a seasonal variation with higher values occurring in summer and lower values in spring and autumn. N_2O fluxes from the estuaries were usually higher than those from the adjacent marine area (Table 4), except August and October 2006. But in September 2003, sea to air N_2O fluxes from the Changjiang Estuary were found to be much higher than those from the adjacent marine area (Zhang et al., 2008). This difference may be caused by the variation of freshwater discharge. In summer of 2006, the water discharge of the Changjiang was relatively low and the influence of N_2O-rich freshwater was rather limited. For example, the water discharge of August and October 2006 is 38% and 55% lower than the long term monthly average water discharge. The average annual N_2O flux from the Changjiang Estuary was 6.8±3.7, 13.3±7.2 and 14.9±8.3 $\mu mol\, m^{-2}\, d^{-1}$ using LM86, W92 and RC01 relationship, respectively. The annual sea to air N_2O flux from its adjacent marine area was estimated to be 8.5±7.8, 15.3±13.5 and 17.4±15.7 $\mu mol\, m^{-2}\, d^{-1}$ using LM86, W92 and RC01 relationship, respectively. Based on the annual sea to air N_2O fluxes resulted from all models, the annual mean atmospheric N_2O fluxes for the Changjiang Estuary and its adjacent marine area were 11.6±4.3 and 13.7±4.7 $\mu mol\, m^{-2}\, d^{-1}$, respectively. These results are comparable to the flux of 8.4–12 $\mu mol\, m^{-2}\, d^{-1}$ reported for Tamar estuary (Law et al.,

BGD
7, 3125–3151, 2010

1992), but are much lower than the flux of 43 200 μmol m^{-2} d^{-1} reported for Humber estuary (Barnes and Owens, 1998) and 265.6±280.4 μmol m^{-2} d^{-1} for Colne estuary (Dong et al., 2004). This study indicated that N_2O fluxes may vary greatly in different estuarine systems. Owing to this great spatial variation, more observations at typical estuarine systems in the world should be done to understand the estuarine N_2O emissions on a global scale. In any way, the Changjiang Estuary and its adjacent marine area act as a significant source of atmospheric N_2O.

The area of the Changjiang Estuary was mainly limited to 122–123° E and 29–32° N and estimated to be ~3.4×10^4 km^2, and its adjacent marine area was limited to the rest area between 122–125° E and 28–34° N and estimated to be ~16.9×10^4 km^2. Considering the estimated area and the corresponding annual mean atmospheric N_2O fluxes for the Changjiang Estuary and its adjacent marine area, the annual N_2O emission from the studied region were estimated to be 6.1×10^8 mol yr^{-1} by the LM86 equation, 11.1×10^8 mol yr^{-1} by the W92 equation and 12.6×10^8 mol yr^{-1} by the RC01 equation.

4 Conclusions

Dissolved N_2O concentrations in the Changjiang Estuary and its adjacent area fall within but toward the lower end of the N_2O range in the worldwide estuaries, and they showed obvious seasonal variation with the high values occurring in summer and spring.

Input via rivers act as a significant source of N_2O in the estuarine water of Changjiang Estuary and its adjacent area. The average annual input of N_2O from the Changjiang to the Estuary and its adjacent area was estimated to be 15.8×10^6 mol/yr. Sediment can act as either a source or a sink of N_2O in the water column of the Changjiang Estuary, however more measurements on the N_2O emission rates at different seasons in Changjiang Estuary are required to assess the source strength of sediment release.

BGD
7, 3125–3151, 2010

The Changjiang Estuary and its adjacent area act as a significant source of atmospheric N_2O. The annual N_2O emissions from the studied region were estimated to range from 6.1×10^8 to 12.6×10^8 mol yr^{-1}. Due to the great spatial variation of N_2O fluxes from different estuaries, more studies on typical estuarine systems in the world are required in the future to estimate the estuarine N_2O emissions accurately on a global scale.

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BGD

7, 3125–3151, 2010

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BGD
7, 3125–3151, 2010

Table 1. Observed N_2O concentrations in the surface and bottom waters of the Changjiang Estuary and its adjacent area. (Numbers in the parentheses are the saturations of N_2O in %.)

		Surface N_2O (nM)		Bottom N_2O (nM)	
Date	Stations	Range	Mean ± S.D.	Range	Mean ± S.D.
25 Apr—3 May 2002	28	8.17–18.9 (103–221)	11.77±2.15 (141±23)	8.49–14.3 (108–187)	12.0±1.34 (147±17)
4–11 Nov 2002	30	6.59–16.7 (84–200)	9.00±2.11 (114±25)	5.83–18.2 (81–220)	9.38±2.13 (125±26)
2-11 June 2006	25	7.48–10.68 (110–135)	9.10±0.88 (119±6)	9.28–16.10 (110–203)	11.03±1.87 (138±26)
15–31 Aug 2006	30	6.04–21.32 (101–363)	10.78±4.25 (184±71)	6.67–20.72 (70–259)	9.66±3.13 134±44
3–13 Oct 2006	30	6.90–13.49 (106–201)	10.11±1.56 (153±24)	6.75–18.44 (99–245)	11.80±2.42 (169±32)

BGD
7, 3125–3151, 2010

Table 2. Summary of N_2O measurements in various estuaries in the literatures.

Study Area	Stations	Date	Sur. N_2O, (nM)	Sur. R, (%)	Salinity	Flux ($\mu mol\, m^{-2}\, d^{-1}$)	Ref.
Danube estuaries	35	Jul–Aug 1995	(8.0±1.0)	(112)		(1.6[a]); (2.8[b])	Amouroux et al. (2002)
Rhone River plume	9	June 1998	16.76–41.04				Marty et al. (2001)
Tamar estuary	18*	Aug 1988	9–30[c]		0–30[c]	(12)	Law et al. (1992)
	29[c]	Oct 1988	9–24[c]		0–30[c]	(11)	
	22[c]	Mar 1989	15–23[c]		0–20[c]	(11)	
	22[c]	Jun 1990	10–18[c]		0–30[c]	(8.4)	
Humber estuary	11[c]	Mar–Dec 1996		100–4250 (452)	0–30		Barnes and Owens (1998)
Tweed estuary		Sep 1996–Mar 1997		96–110 (100.4)			
Gironde estuary	9	Nov 1991	(14.3)	106–165 (132)	5–34		Bange et al. (1996)
		Jun 1997	9.8–36.7		0–30		De Bie et al. (2002)
Loire estuary	13	Sep 1998	7.3–21		0–32		De Bie et al. (2002)
Schelde estuary	15	Oct 1978	10–250	120–3000(1560)	0–30		de Wilde and de Bie (2000)
	35[c]	Oct 1993	10–338		2–30		
	22[c]	Mar 1994	75–150		2–10		
		Jul 1996	50–300		2–22		
Schelde estuary	10	May 1997–Apr 1998	8.7–1457	(710)	0–25		de Bie et al. (2002)
Thames estuary	15	Feb 1998	11.2–93	93–681(321)	0–33		
Childs estuary		Jun–Jul 1993	2–38[c]	(302)			LaMontagne et al. (2003)
		Nov–Dec 1992	4–65[c]	(613)			
Colne estuary		Aug 2001	94.1	761		137.0	Dong et al. (2004)
		Nov 2001	90.6	538		101.3	
		Feb 2002	132.5	450		138.8	
Colne estuary		May 2002	467.5	2187		685.5	Robinson et al. (1998)
Seine River estuary		Summer				39–98	Garnier et al. (2007)
Pearl River estuary	14	Sep 2003	20.4–48.6		0–5		Chen et al. (2008)
	1	Apr 2004	62.6	720			Xu et al. (2005)
Temmesjoki estuary	3	Summer 2004	7.3–15	136		5–7	Silvennoinen et al. (2008)

[a] K_w was estimated by the LM86 equation;

[b] K_w was estimated by the W92 equation;

[c] Data was estimated from the reference.

Numbers in the parentheses are the average value.

BGD

7, 3125–3151, 2010

Table 3. Compilation of dissolved N_2O in various rivers. (Numbers in the parentheses are the averaged value.)

Rivers	Description	N_2O (nM)	N_2O (%)	Ref.
Alsea River	Oct 1979	8.2–15.6	94–166	De Angelis and Gordon (1985)
South Platte river	Jan/Feb 2000	18–527 (59)	2500	Dennehy and McMahon (2000)
Arkansas River	Jan/Feb 2000	3.3–5.9 (3.6)		
Potomac River	Jul/Sep 1977	7–350	100–5000	McElroy et al. (1978)
Hudson River	1998–1999, monthly measurement	(19±8)	125–385 (185±43)	Cole and Caraco (2001)
English rivers				Dong et al. (2004)
Colne	Aug 2001–May 2002	(44.2±5.0)	(272.5±32.1)	
Ouse	Aug 2001–May 2002	(39.2±2.9)	(217.9±16.5)	
Trent	Aug 2001–May 2002	(43.2±3.5)	(228.4±18.3)	
Stour	Aug 2001–May 2002	(53.9±4.5)	(297.4±25.7)	
Orwell	Aug 2001–May 2002	(60.1±5.4)	(389.1±31.7)	
Amazon River	Mainstem	(13.4±2.5)		Richey et al. (1988)
Millstone River	Mar–Ma 2002	11.45–13.30	104–123	Laursen and Seitzinger (2004)
Iroquois River	Apr–Jun 2002	13.65–27.67	134–209	
Changjiang	Aug 2002	9.02±1.03		Yan et al. (2004)
	Oct 2002	13.39±8.71		
	Jan 2008	22.0±3.5	168±27	Zhao et al. (2009)
Pearl River	Sep 2003	20–40		Chen et al. (2008)
	Apr 2004	62–323 (143)	720–4080 (1730)	Xu et al. (2005)
River Temmesjoki	2003–2004	6.7–53 (19)		Silvennoinen et al. (2008)

BGD
7, 3125–3151, 2010

Table 4. N_2O fluxes from the Changjiang Estuary and its adjacent marine area (LM86: K_w was calculated using the tri-linear k/u_{10} relationship established by Liss and Merlivat (1986); W92: K_w was calculated using the quadratic k/u_{10} relationship established by Wanninkhof (1992); RC01: K_w was calculated using the k/u_{10} relationship established by Raymond and Cole (2001).

Region	Seasons	*n*	Surface N_2O (nM)	Surface N_2O (%)	ΔC (nM)	U_{10} (m/s)	*F* (LM86) (μmol m^{-2} d^{-1})	*F* (W92) (μmol m^{-2} d^{-1})	*F* (RC01) (μmol m^{-2} d^{-1})	Mean Flux (μmol m^{-2} d^{-1})
Estuary (*s*<30)	May 2002	9	13.32±2.32	153±28	4.61±2.39	6.7	9.0±4.7	17.5±9.0	19.0±9.8	15.2±5.4
						(5.9±2.2)[a]	6.3±5.3	14.1±10.6	17.7±15.6	
	Jun 2006	11	9.37±0.50	120±5	1.52±0.42	6.8	3.3±0.9	6.4±1.8	7.0±1.9	5.6±2.0
	Aug 2006	11	9.67±3.27	161±54	3.65±3.24	7.6	11.8±10.5	23.3±20.6	26.9±23.8	20.7±7.9
	Oct 2006	3	9.11±1.47	135±25	2.33±1.64	7.2	6.2±4.5	12.2±8.8	13.6±9.9	10.7±3.9
						(3.1±2.2)[a]	1.6±2.6	3.0±4.5	4.5±5.7	
	Nov 2002	16	9.59±2.45	117±31	1.35±2.52	7.6	3.5±6.5	6.9±12.7	8.0±14.7	6.1±2.3
	Annual mean		10.21±1.75	137±20	2.69±1.41	7.2±0.4	6.8±3.7	13.3±7.2	14.9±8.3	11.6±4.3
Marine area (*s*>30)	May 2002	15	10.85±1.44	134±16	2.75±1.29	6.7	5. 5±2.5	10.6±4.9	11.5±5.4	11.1±0.6
						(4.7±1.7)[a]	2.6±3.3	6.1±6.4	7.3±8.3	
	Jun 2006	14	8.90±1.06	118±6	1.39±0.54	6.8	3.0±1.1	5.8±2.1	6.4±2.3	5.1±1.8
	Aug 2006	19	11.42±4.69	198±78	5.69±4.60	7.6	18.9±15.0	37.1±29.5	42.9±34.1	33.0±12.5
	Oct 2006	26	10.22±1.56	156±24	3.65±1.56	7.2	9.8±4.2	19.1±8.1	21.4±9.1	16.8±6.1
						(6.2±2.6)[a]	7.8±7.3	14.1±12.3	22.6±23.0	
	Nov 2002	11	8.13±1.08	111±28	0.77±0.94	7.6	2.1±2.5	4.0±4.9	4.7±5.7	3.6±1.3
	Annual mean		9.90±1.36	143±35	2.85±1.95	7.2±0.4	8.5±7.8	15.3±13.5	17.4±15.7	13.7±4.7

[a] Numbers in the parentheses are the ship-based in situ wind speeds.

BGD
7, 3125–3151, 2010

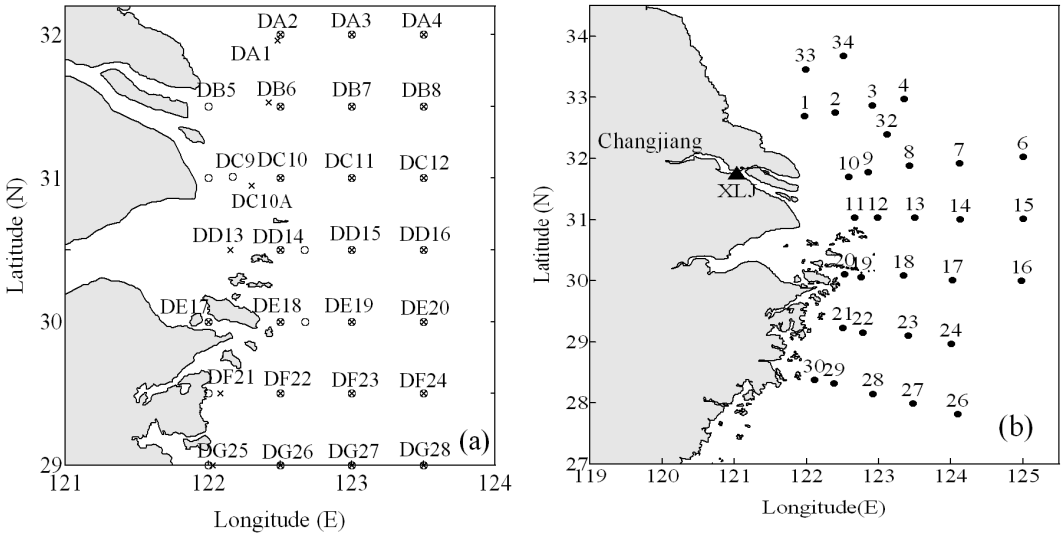

Fig. 1. Sampling locations in the Changjiang (Yangtze River) Estuary. **(a)** April (×) and November (o) of 2002; **(b)** June, August, October of 2006 (•) and Xuliujing (XLJ, solid triangle).

BGD
7, 3125–3151, 2010

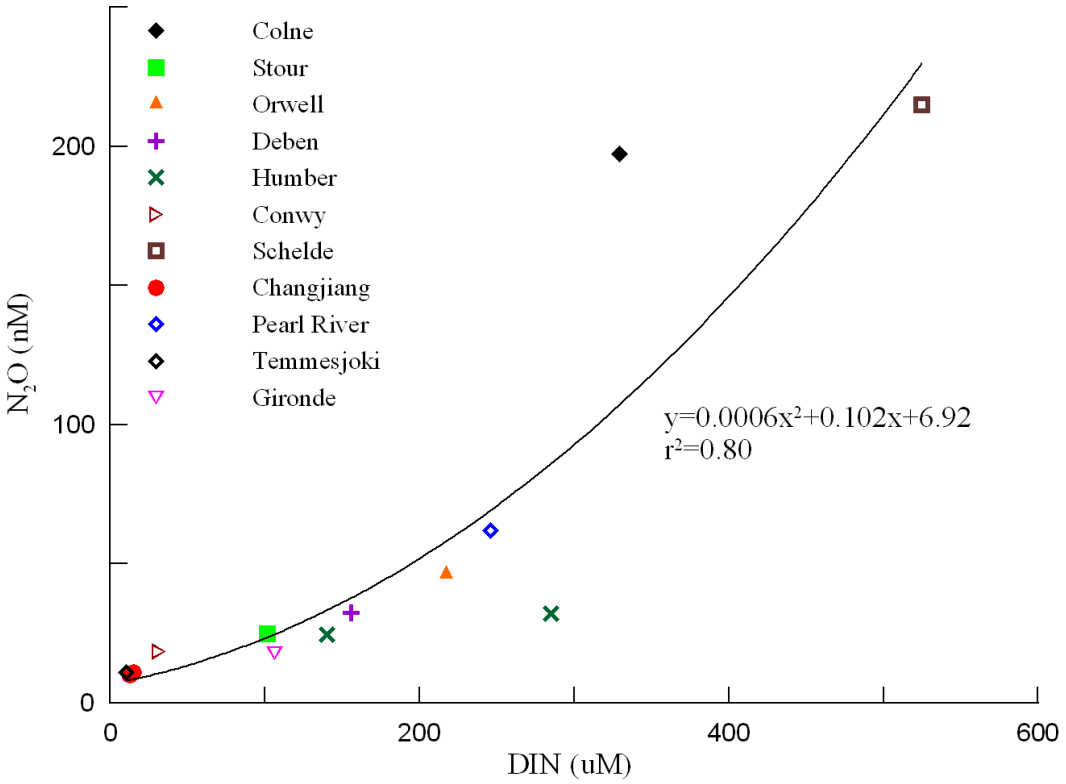

Fig. 2. N_2O concentrations versus DIN for various estuaries. (The data were from this study and the references in Table 2.)

BGD
7, 3125–3151, 2010

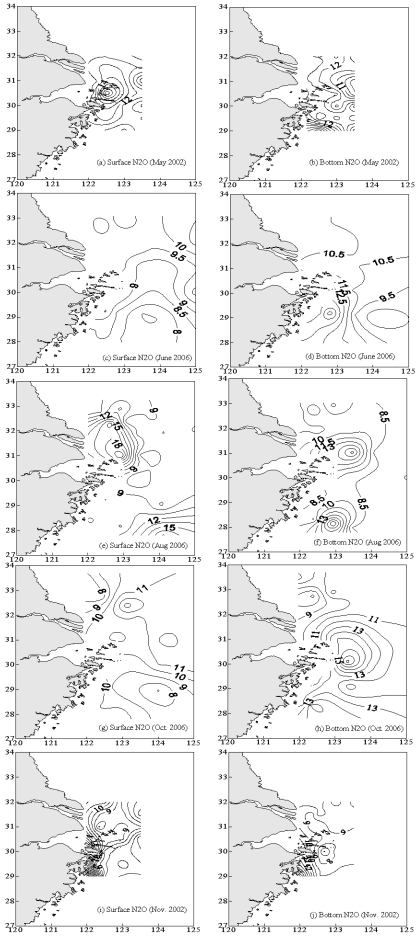

Fig. 3. Horizontal distributions of N_2O (nM) in the surface and bottom waters of the Changjiang Estuary.

BGD
7, 3125–3151, 2010

Nitrous oxide in the Changjiang Estuary and its adjacent marine area

G.-L. Zhang et al.

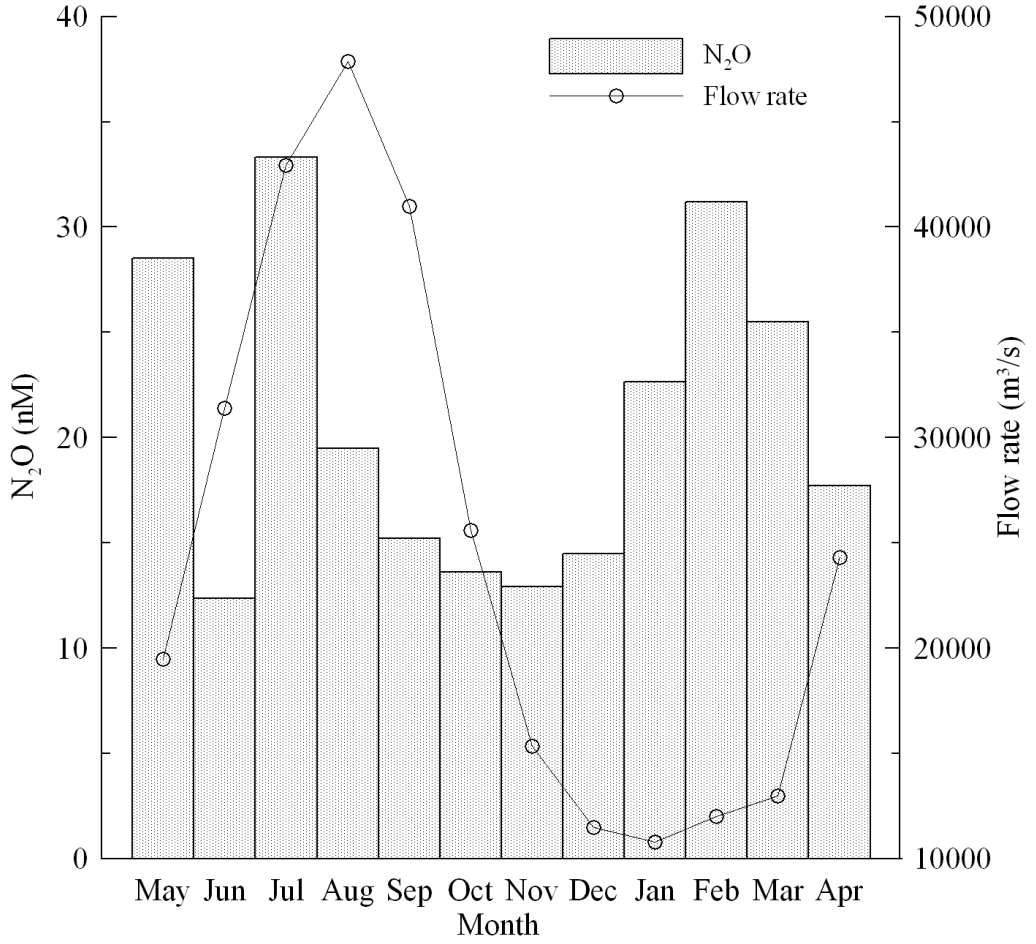

Fig. 4. Monthly variation of N_2O concentrations at Xuliujing and flow rates at Station Datong in the Changjiang from May 2007 to April 2008.

BGD
7, 3125–3151, 2010