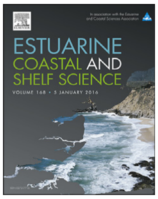
Estuarine, Coastal and Shelf Science 168 (2016) 22–28

Contents lists available at ScienceDirect

Estuarine, Coastal and Shelf Science

journal homepage: www.elsevier.com/locate/ecss



Distributions and fluxes of nitrous oxide in lower reaches of Yellow River and its estuary: Impact of water-sediment regulation

Xiao Ma [a, b, 1], Gui-Ling Zhang [a, b, *], Su-Mei Liu [a, b], Lan Wang [a], Pei-Pei Li [a], Pei-Pei Gu [a], Ming-Shuang Sun [a, 1]

[a] Key Laboratory of Marine Chemistry Theory and Technology, Ministry of Education, Ocean University of China, 266100 Qingdao, PR China
[b] Qingdao Collaborative Innovation Centre of Marine Science and Technology, Ocean University of China, 266100 Qingdao, PR China

ARTICLE INFO

Article history:
Received 12 March 2015
Received in revised form
14 September 2015
Accepted 8 October 2015
Available online 14 November 2015

Regional index terms:
China
Shandong Peninsula
Yellow River (Huanghe) estuary

Keywords:
River water
Sediments
Nitrous oxide
Distribution
WSR: water-sediment regulation

ABSTRACT

The Yellow River is the second largest river in China and is well-known for its high sediment load. Since 2002, water-sediment regulation has been performed annually to scour the silted river channel in the lower reaches and to promote release of sediment from the large reservoirs. Here we present a comprehensive study of the variations in distribution and emission of dissolved N_2O in the lower reaches of the Yellow River and its estuary based on: *(i)* monthly sampling at a lower-river station (Kenli) from November 2008 to December 2009, *(ii)* daily monitoring at this station during the water-sediment regulation event in summer of 2009, and *(iii)* field surveys of the estuary before, during, and after the regulation event. N_2O concentrations in the lower reaches of the Yellow River ranged from 8.78 to 24.26 nmol/L, and had high values in winter and spring. N_2O flux from the Yellow River to the Bohai Sea was about 2.27×10^5 mol/year. Water-sediment regulation had a strong impact on N_2O distribution and transportation in the lower Yellow River. A sharp increase of N_2O (8-fold) occurred at the beginning of water-sediment regulation, and this excessive N_2O was likely from stimulation of nitrification in the water column. A total of 55.9% of the annual N_2O input from the Yellow River to the Bohai Sea occurred during water-sediment regulation, but the corresponding water discharge during this period accounted for only 26.9% of total runoff. N_2O concentrations in the lower reaches of the Yellow River and its estuary were almost all super-saturated, and this region acted as a net source of atmospheric N_2O. High N_2O saturations and air-sea fluxes were present in the Yellow River estuary during water-sediment regulation. These results indicate that water-sediment regulation in the Yellow River has a great impact on the estuarine distribution and atmospheric emission of N_2O, and that this effect lasts for several weeks.

1. Introduction

Nitrous oxide (N_2O) is the third most significant greenhouse gas, only after carbon dioxide (CO_2) and methane (CH_4) (Hartmann et al., 2013). Although the atmospheric mixing ratio of N_2O is several orders of magnitude lower than that of CO_2 and CH_4, N_2O has a strong effect on global warming because its long atmospheric lifetime (131 years, Prather et al., 2012) and high global warming potential (Myhre et al., 2013). Since the industrial revolution, the atmospheric mixing ratio of N_2O has risen from 270 ppbv to 324.2 ppbv, corresponding to about 0.75 ppbv/year (Hartmann et al., 2013). Anthropogenic sources, such as farm soils, rivers and estuaries, fossil fuel combustion, and industrial discharge, are the main causes of the increasing atmospheric N_2O (Montzka et al., 2011; Rao and Riahi, 2006; Ciais et al., 2013). Rivers and estuaries often undergo eutrophication due to excessive nitrogen input from fertilizers, discharge of sewage and waste, and atmospheric deposition (Howarth et al., 2011; Voss et al., 2011). Thus, even though they account for a relatively small amount of the earth's surface, rivers and estuaries are significant sources of atmospheric N_2O. The latest IPCC report estimated that global N_2O emission from rivers, estuaries and coastal zones could be as large as 0.6 Tg N/year (Ciais et al., 2013), thus accounting for about 3% of global N_2O emissions.

The Yellow River (Huanghe), which is the second longest river in China, has an extremely high load of sediment from the Loess

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[*] Corresponding author. Key Laboratory of Marine Chemistry Theory and Technology, Ministry of Education, Ocean University of China, 266100 Qingdao, PR China.

E-mail address: guilingzhang@ouc.edu.cn (G.-L. Zhang).

[1] Present address: Helmholtz Centre for Ocean Research Kiel (GEOMAR), Kiel, Germany.

http://dx.doi.org/10.1016/j.ecss.2015.10.001
0272-7714/

Plateau. It flows east for 5164 km and empties into the Bohai Sea, draining a basin of 752,443 km^2 (Yu, 2006). The Yellow River was once considered the second largest river in terms of sediment load; however, recent research estimated the sediment load was less than 100 Mt/year due to increasing water consumption and drought (Milliman and Farnsworth, 2011). Nonetheless, significant soil erosion and heavy sand yield have led to exceptional transport of sediment toward the sea and excessive sediment deposits at lower reaches. This has caused silting of the river channel and a growing riverbed, which may be several meters above the ambient level.

The Yellow River Conservancy Commission has enforced water-sediment regulation (WSR) since 2002 (Yu, 2006). Artificial flood peak discharge was created for 20–35 days each year to remove silt from the river channel in the lower reaches and to release sediment from the large reservoirs by the joint operation of Xiaolangdi and two other reservoirs. WSR was implemented from June 19 to July 8 in 2009, during which 3488 million m^3 water and 34.3 million tons of sediment were flushed into the Bohai Sea (Yellow River Sediment Bulletin, 2009).

Abrupt increases of water sediment and its transportation have had profound physical, ecological, and geomorphologic effects on the riverine and estuarine areas of the Yellow River (Wang et al., 2007; Fan and Huang, 2008; Liu et al., 2012). Most research on the Yellow River has focused on nutrients and carbon cycles (Liu et al., 2012, 2014), and little is known about the N_2O yield and transportation. The unique character of the water-sediment of the Yellow River distinguishes it from other major rivers in the world. The implementation of the WSR provides an excellent case for study of an altered river system that has been affected by extensive anthropogenic activities. Here we present a study of dissolved N_2O in the lower reaches of Yellow River, with a focus on variations in N_2O concentrations and the impact of WSR on riverine and estuarine N_2O distribution and emission.

2. Materials and methods

2.1. Sample collections

Surface water samples were collected monthly from November 2008 to December 2009 at the Kenli station in the lower reaches of the Yellow River, about 72 km upstream of the river mouth (Fig. 1). River water was transferred from a sampling bucket to 56.5 mL glass vials, and 0.5 mL of a saturated $HgCl_2$ solution was then added. Bubble-free samples were sealed immediately with butyl rubber stoppers and aluminum caps. For investigation of the spatial distribution of dissolved N_2O along the lower reaches, samples were also collected in November 2008 and June 2009 at stations in Lijin, Jianlin and Qingba, which are about 94 km, 49 km, and 14 km upstream from the river mouth, respectively.

Daily observations (8–9 AM) were conducted at the Kenli station during the WSR event from June 19 to July 18 in summer of 2009, but there was no sampling on July 1 and 2 due to fierce torrents. Water discharge data were collected from the Lijin hydrological station (about 22 km upstream of the Kenli station), and there are no branch inputs or significant agriculture outputs between these stations. In addition, three surveys, before WSR (June 15), during WSR (July 1), and after WSR (July 19), were performed at the Yellow River estuary and its adjacent area (Fig. 2). Surface water samples were collected and temperature and salinity (using the Practical Salinity Scale) were obtained from the shipboard conductivity, temperature, and depth (CTD) profiles. Atmospheric pressure and wind speed were collected by an anemometer (AZ8910, China).

2.2. Chemical analysis

Dissolved N_2O was measured with a gas chromatograph (GC-14B, Shimadzu) using the head-space equilibrium method (Walter et al., 2006). Electron capture detector (ECD) response toward N_2O was calibrated with different standard gases (330 ppbv, 380 ppbv, and 5000 ppbv N_2O/N_2, Research Institute of China National Standard Materials). The detection limit for N_2O was 1.0 nmol/L, and the precision was about 2%. Nutrients were measured photometrically in the laboratory by an auto-analyzer (Model: Skalar SAN^{plus}), which had precision less than 5–10%.

2.3. Computation of sea-to-air fluxes

Sea-to-air flux was calculated as:

$$F = k \times (C_{obs} - C_{eq}) \tag{1}$$

where C_{obs} is the measured concentration of a dissolved gas in the surface seawater, C_{eq} is the air-sea equilibrated concentration (which can be calculated from *in situ* temperature and salinity), and k is the gas transfer velocity. Atmospheric N_2O concentration was not measured directly in this study, and a global mean of 323 ppbv for 2008 and 2009, from the NOAA/ESRL Global Monitoring Division *in situ* program (http://www.esrl.noaa.gov/gmd), was used for calculations. The gas transfer velocity (k) is a function of wind speed and the Schmidt number. The Schmidt number is calculated by empirical equations with seawater kinematic viscosity and the diffusion coefficient of N_2O (D_{N2O}) in water (Walter et al., 2004).

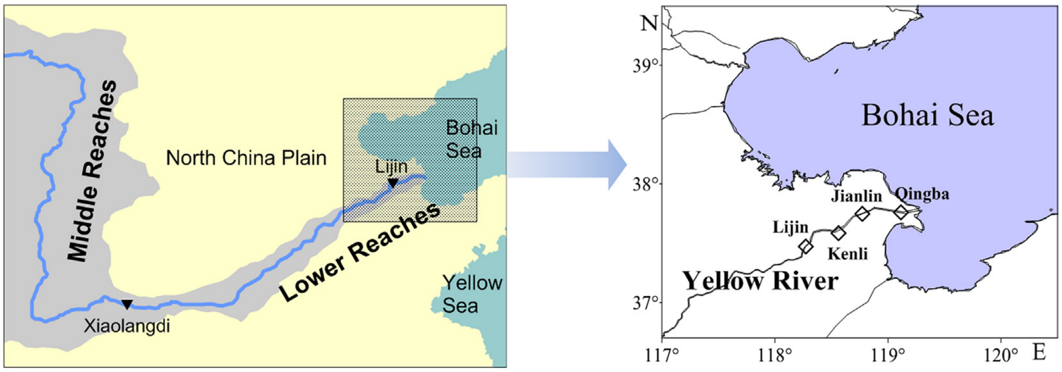

Fig. 1. Sample locations (◊) on the Yellow River.