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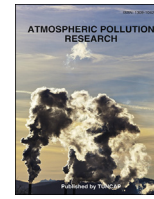

Temporal and spatial variability of ground level atmospheric methane concentrations in the Ebro River Delta

Alba Àgueda [a, *], Claudia Grossi [a, **], Elsa Pastor [b], Eva Rioja [a, b], Laura Sánchez-García [a, 1], Òscar Batet [a], Roger Curcoll [a], Marina Ealo [a, 2], Manel Nofuentes [a], Paola Occhipinti [a], Xavier Rodó [a, c], Josep-Anton Morguí [a, d]

[a] *Catalan Institute of Climate Sciences (IC3), Barcelona, Spain*

[b] *Centre for Technological Risk Studies (CERTEC), Department of Chemical Engineering, Universitat Politècnica de Catalunya·BarcelonaTech, Barcelona, Spain*

[c] *Catalan Institution for Research and Advanced Studies (ICREA), Barcelona, Spain*

[d] *Ecology Department, Universitat de Barcelona (UB), Barcelona, Spain*

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A B S T R A C T

Deltas provide many worthy ecosystem services. Yet, delta basins are quite vulnerable, especially in the face of climate change, which can affect the outcome of both agriculture and biodiversity. Moreover, rice paddy cultivation is well known to contribute with strong emissions of greenhouse gases (GHGs), such as methane (CH_4). Thus, knowing the atmospheric variability of CH_4 in relation to the different stages of the rice culture cycle could help to improve GHGs' mitigation strategies in deltas.

The Ebro River Delta, in the northwestern Mediterranean basin, forms part of the largest Spanish river basin and is mainly covered by rice fields. In this study, for the first time, ground level (40 cm a.g.l.) atmospheric CH_4 concentrations have been monitored in this area, through twenty-seven car mobile transects, over the course of one year. Seasonal, diurnal and spatial variability of CH_4 concentrations were studied to identify its relationship with rice cultivation, meteorological conditions and land-use distribution.

With regard to seasonal variability, autumn transects showed the highest mean values for atmospheric CH_4 (2.466 ppm) when dead rice straw is mixed with the sediment, and weed growth is prevented. Spring and summer measurements gave the highest mean CH_4 values at dawn (1.897–3.544 ppm), whereas autumn and winter produced the mean values after sunset (2.148–2.930 ppm). Spatial differences were accounted for by proximity to urban areas, presence of shallow water storage structures, and distance to seawater.

1. Introduction

Methane (CH_4) contributes greatly to the natural and anthropogenic greenhouse gas effect. Globally averaged surface CH_4 concentrations have risen from 722 ± 25 ppb in 1750–1803 ± 2 ppb by 2011, and this increase has been linked predominantly to human activities (IPCC, 2013).

Sources of CH_4 have been identified to be of three main types: (1) thermogenic: seepages, mud volcanoes, geothermal vents, leakages from fossil fuel extraction and use; (2) pyrogenic: incomplete combustion of fossil fuel and plant biomass; (3) biogenic: wetlands, termites, oceans, rice paddies, ruminant livestock, landfills, etc. (IPCC, 2013). The most important sinks of CH_4 include its tropospheric and stratospheric oxidation by OH· radicals and its microbial oxidation in methanogenic soils and upland soils.

According to IPCC (2013) estimates, natural wetlands and rice paddies contribute about 37.3% (32% and 5.3%, respectively) of the

* Corresponding author.
** Corresponding author.
E-mail addresses: alba.agueda@ic3.cat (A. Àgueda), claudia.grossi@ic3.cat (C. Grossi).

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[1] Present address: Centro de Astrobiología (CSIC-INTA), Madrid, Spain.
[2] Present address: Institute of Environmental Assessment and Water Research, CSIC, Barcelona, Spain.

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CH_4 emitted into the atmosphere, while upland soils are responsible for 4.4% of the CH_4 consumption. These values change a little if Spanish inventory data are taken into consideration. Spanish rice paddies contribute only about 1% of the net CH_4 emitted by this country, while values for wetlands and upland soils are not reported (Ministerio de Agricultura and Alimentación y Medio Ambiente, 2016). Although main soil related sources and sinks of CH_4 are well identified, their relative contributions to atmospheric CH_4 levels are still highly uncertain (Kirschke et al., 2013).

Flooded rice paddies and wetland environments have a predominantly oxygen-free (anoxic) soil profile. In these ecosystems, CH_4 is produced by methanogenic bacteria that digest organic matter under anaerobic conditions (methanogenesis) (Zhang et al., 2011). CH_4 can also be oxidized into CO_2 by methanotrophs in aerobic zones (e.g. rhizosphere, lower part of culms, soil-water interface and submersion water) (Le Mer and Roger, 2001). Atmospheric CH_4 concentrations measured in the lower boundary layer of these ecosystems are the result of the transfer by diffusion, ebullition or mostly through aerenchyma of the net CH_4 produced at the soil-water/soil-atmosphere interface, as well as the result of atmospheric mixing.

In recent years, there have been many studies on the CH_4 emitted from rice fields. Several factors that control these emissions have been highlighted, including both environmental and agricultural factors. To mention only a few: soil and air temperature (e.g. Pereira et al., 2013; Chanton et al., 1997; Khalil et al., 1998b; Simpson et al., 1995), soil redox potential (e.g. Oo et al., 2015; Simpson et al., 1995), water management (e.g. Meijide et al., 2011; Yan et al., 2005; Miyata et al., 2000), organic amendment (e.g. Yan et al., 2005; Fitzgerald et al., 2000) and fertilizers management (e.g. Oo et al., 2015; Linquist et al., 2012; Bodelier et al., 2000). Most of those studies consider only CH_4 emissions during the crop cycle, and very few attempts have been made to measure CH_4 emissions during the fallow periods of rice soils (e.g. Fitzgerald et al., 2000; Bronson et al., 1997), although straw management (e.g. incorporation into the field, removal from the field, or burning) and flooding practices after harvest can substantially affect CH_4 emissions (e.g. Fitzgerald et al., 2000).

Moreover, these studies have been performed in specific plots of research stations located in the USA (Chanton et al., 1997) and Japan (Miyata et al., 2000), or directly in individual rice paddies located in China (Khalil et al., 1998b), Philippines (Simpson et al., 1995), the USA (Fitzgerald et al., 2000), Italy (Meijide et al., 2011), Portugal (Pereira et al., 2013) and Myanmar (Oo et al., 2015). Thus, to date little effort has been devoted to assessing CH_4 emissions in flooded rice field areas in Europe.

Although continuous atmospheric CH_4 observations are currently being performed at several worldwide existing measurement sites to improve our understanding of CH_4 concentration trends at the regional and continental level (e.g. InGOS - http://www.ingos-infrastructure.eu/ - and ICOS - https://www.icos-ri.eu/ - European projects), spatial CH_4 mobile surveys have only been recently performed to identify areas of high CH_4 concentrations, and link them to specific sources. Research studies where CH_4 transects have been executed have usually focused on surveying long distances covered by different land-uses (Bamberger et al., 2014; Farrell et al., 2013), cities (Zazzeri et al., 2015; Phillips et al., 2013; Shorter et al., 1996) or natural gas and oil production sites (Eapi et al., 2014; Pétron et al., 2012), where localized sources can be identified. In contrast, spatial differences within rice field dominated areas have rarely been considered (e.g., Oo et al., 2015; Mukherjee et al., 2012).

In this study the Ebro River Delta (ERD), a rice paddy dominated region in the northwestern Mediterranean basin, has been surveyed twenty seven times (9 days) by car, over the course of a year (December 2011–November 2012). The ERD is of international importance due to its abundant flora and fauna, and belongs to the largest river basin in Spain. The car surveys were executed during different phases of the diurnal-seasonal cycles to understand the temporal and spatial variation of ground level CH_4 concentrations in relation to atmospheric conditions and agricultural practices. We consider that improving knowledge of CH_4 concentration levels in natural areas of the Mediterranean basin is very important given that it is a vulnerable region in terms of climate change. In addition, since this area is dominated by rice paddies, the results of this study could offer useful information to help in the improvement of the agricultural Best Management Practices for CH_4 emissions reduction.

2. Material and methods

2.1. Study site: Ebro River Delta

The present study was carried out in the ERD, in northeastern Spain (Fig. 1). The ERD is one of the largest wetland areas (over 300 km^2) in the western Mediterranean region and is located on a relatively small peninsula.

The Ebro Deltaic area has a typical Mediterranean climate with mild winters and warm summers (Casanova, 1998). Drought is observed in summer and rainfall is usually concentrated in two different periods of the year (March–May and September–November). Wind blows with high mean annual velocities (>30 km h^{-1}) during the entire year (Generalitat de Catalunya, 2011), and blows predominantly from the NW in winter (e.g. Casanova, 1998; Grossi et al., 2016), and from the S-SE in summer (Generalitat de Catalunya, 2011). The atmospheric relative humidity is high over the entire year (>65%) (e.g. Generalitat de Catalunya, 2011; Grossi et al., 2016).

The ERD has a flat orography. Maximum elevations are found near the town of Amposta (8 m above sea level (a.s.l.)) and close to the river, with values of only 4–5 m a.s.l. (Generalitat de Catalunya, 2011). About 60% of the whole area has elevations lower than 1 m a.s.l. (Generalitat de Catalunya, 2011). Sediment supply to the deltaic plain has been reduced during recent years, with the result that the vertical accretion process in the deltaic plain has been limited (Alvarado-Aguilar et al., 2012).

The ERD has a shallow aquifer (6–8 m deep) formed of low-permeability fine sand, allowing the appearance of frequent saline and hypersaline zones (Bayó et al., 1993). A deep aquifer (40–60 m) also exists and is present in several layers (gravels, sand, silts), but it has been poorly characterized so far (Bayó et al., 1993). The ERD is irrigated with surface water taken from the Ebro river 56 km upstream. Two main canals distribute water through a network of smaller canals at both margins of the river.

The dominant land-use in the ERD is rice agriculture. Rice paddies are spread over the major part of the Delta (Fig. 1, zoom in panel) and represent 62% of the cultivated area (Fatoric and Chelleri, 2012). As there is a saline environment at the upper soil level, continuous flooding allows rice to be grown in the Delta, because this type of plant can get oxygen through the aerenchyma when flooded, and salt levels are lowered.

Shallow coastal waters, beaches, dunes, saline lagoons, salinas, freshwater marshes, and freshwater pools fed by groundwater springs are also present in this fluvial Delta. Most of the natural wetland areas are found parallel to the coastline (Fig. 1, zoom in panel) and within the limits of the Natural Park, declared a protected area in 1983. The ERD is included in the List of Wetlands of International Importance (RAMSAR, 2016) and supports numerous species of waterbirds, commercially valuable fish, mollusc, shrimp and endemic fish populations.

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