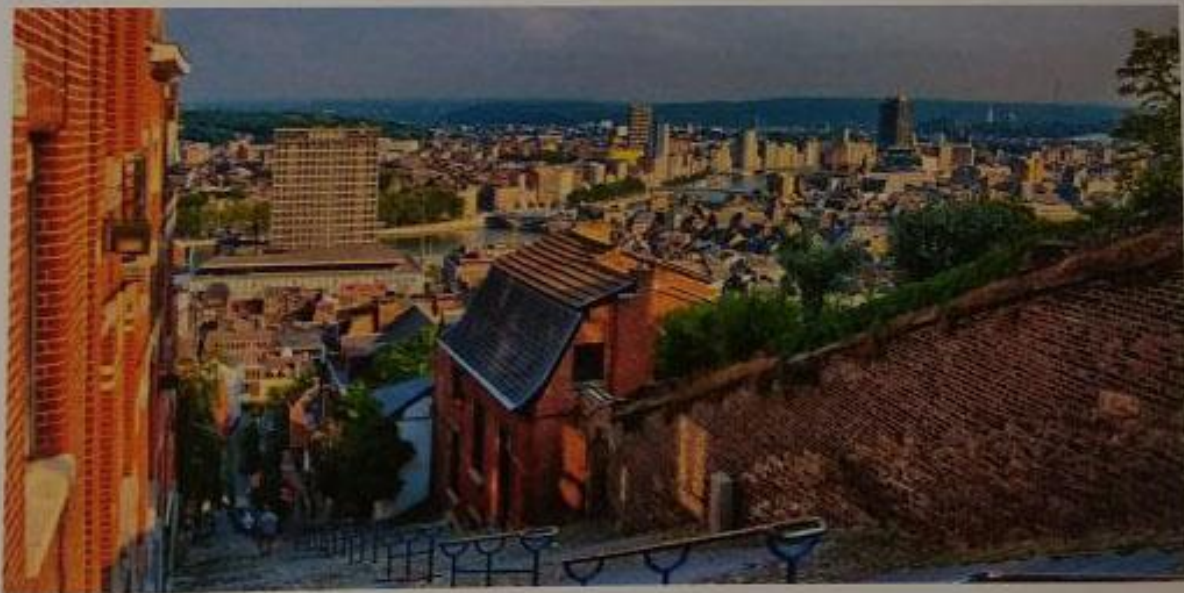


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+32 478 50 66 84

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www.basis-online.eu
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Benelux Association of Stable Isotope Scientists "BASIS"
Secretary: Isotope Bioscience Laboratory - Coupure links 653, 9000 Gent, Belgium
Bank: 1226.21.565 Rabo, Wolphaartsdijk, The Netherlands
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Dynamics of nitrous oxide in groundwater under agricultural areas: insights from multi-isotopic studies (^{15}N , ^{34}S , ^{18}O , ^{13}C , ^3H)

1. Olha Nikolenko 2. Philippe Orban 3. Pierre Jamin 4. Anna Jurado 5. Caroline Thomas 6. Cédric Morana 7. Alberto V. Borges 8. Serge Brouyère

1. University of Liège, ArGENCo, Hydrogeology and Environmental Geology, Aquapôle, -B52/3 Sart-Tilman, 4000 Liège, Belgium 2. University of Liège, ArGENCo, Hydrogeology and Environmental Geology, Aquapôle, -B52/3 Sart-Tilman, 4000 Liège, Belgium 3. University of Liège, ArGENCo, Hydrogeology and Environmental Geology, Aquapôle, -B52/3 Sart-Tilman, 4000 Liège, Belgium 4. Department of Hydrosociences, Technische Universität Dresden, Dresden, Germany 5. University of Liège, ArGENCo, Hydrogeology and Environmental Geology, Aquapôle, -B52/3 Sart-Tilman, 4000 Liège, Belgium 6. Chemical Oceanography Unit, University of Liège, Liège, Belgium 7. Chemical Oceanography Unit, University of Liège, Liège, Belgium 8. University of Liège, ArGENCo, Hydrogeology and Environmental Geology, Aquapôle, -B52/3 Sart-Tilman, 4000 Liège, Belgium

In the last two decades the biogeochemistry of nitrous oxide (N_2O), one of the most important greenhouse gases, in the subsurface has started to draw significant research attention due to the rising concern about climate change (Clough et al., 2005). The increase in the concentration of N_2O in the atmosphere reflects the increase in amount of N_2O derived from the anthropogenic sources (Robertson & Vitousek, 2009). Among these sources it is agriculture that contributes nearly 60% of the total anthropogenic emission of N_2O (Syakila & Kroeze, 2011). Agricultural N_2O emission to the atmosphere could be divided into direct (occurring from soils) and indirect (occurring from groundwater and surface water as a result of N input to aquatic systems) contributions. While the former one has been intensively studied and is relatively well constrained, the latter one requires additional investigations (Beaulieu et al., 2011; Jurado et al., 2017). Our study attempts to acquire additional evidence about the N_2O dynamics in the subsurface by studying its distribution across the chalk aquifer of the Geer catchment in Belgium (the area of the basin 480 km^2), where previous studies detected the pronounced impact of the agricultural activities on the groundwater chemistry (Brouyère et al., 2004). To this end, the groundwater samples from 32 locations in confined and unconfined parts of the aquifer were collected in order to examine the spatial variability of N_2O along the lateral and vertical dimensions of the studied aquifer. The results of the study revealed that the concentration of dissolved N_2O in groundwater varied from 0.03 $\mu\text{gN/L}$ to 19 $\mu\text{gN/L}$. The majority of groundwater samples collected in the unconfined part of the chalk aquifer, were supersaturated with N_2O (above 0.3 $\mu\text{gN/L}$), while confined area was characterized with lower values of N_2O concentration (0.02 – 0.12 $\mu\text{gN/L}$). In order to identify the biogeochemical pathways of N_2O , the multiple isotope analysis of NO_3^- , N_2O , SO_4^{2-} , B, DOC and ^3H were conducted. In addition, the dynamics of N_2O production/consumption processes was further explored using data about intramolecular distribution of ^{15}N in N_2O . Analysis of information about isotopic signals of compounds of interest and isotopomer maps of N_2O helps to elucidate the causes of shifting N_2O occurrence in the subsurface. Beaulieu, J. J., Tank, J. L., Hamilton, S. K., Wollheim, W. M., Hall, R. O., Mulholland, P. J., ... & Dodds, W. K. (2011). Nitrous oxide emission from denitrification in stream and river networks. *Proceedings of the National Academy of Sciences*, 108(1), 214-219.

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