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Spatial variability of methane: Attributing atmospheric concentrations to emissions

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ABSTRACT

Atmospheric methane concentrations were quantified along transects in Switzerland, using a mobile laser spectrometer combined with a GPS, to identify their spatio-temporal patterns and their controlling factors. Based on these measurements in complex terrain dominated by agriculture, three main factors were found to be responsible for the diurnal and regional patterns of atmospheric methane: (1) magnitude and distribution of methane sources within the region, (2) efficiency of vertical exchange, and (3) local wind patterns within the complex topography. An autocorrelation analysis of measured methane concentrations showed that nighttime measurements close to the ground provide information about regional sources (up to 8.3 km), while daytime measurements only carry information about sources located up to 240 m away in the upwind fetch. Compared to daytime concentrations, nighttime methane concentrations do also better reflect emissions obtained from a spatially explicit methane emission inventory and allowed the investigation of inconsistencies in this emission inventory.

1. Introduction

The spatial variability of atmospheric methane concentrations and their temporal variations are still insufficiently known, despite the prominent role of methane in atmospheric chemistry, its major contribution (29%) to the positive anthropogenic radiative forcing (IPCC, 2013) and the number of measurement stations in deployment has been steadily increasing in the past (Dlugokencky et al., 2011). With the advent of spatially explicit emission inventories, it becomes increasingly important to find empirical methods for field validation of such inventories. Using an instrumented car for transect measurements in an agriculturally dominated area of Switzerland as done in this study, is a promising technique that is a first step in that direction.

Since conventional measurement systems for methane concentrations and fluxes mostly rely on point measurements at specific sites, spatial interpolation is difficult. Especially in regions with multiple methane sources, the concentrations of atmospheric methane are expected to exhibit significant deviations from the global atmospheric background of 1.80 ppm (NOAA Earth System Research laboratory, http://www.esrl.noaa.gov/gmd/ccgg/figures/) and to give essential information about the emission strength of regional methane sources. Although there are studies which evaluate the regional distribution of methane in the upper atmosphere using aircraft measurements (e.g., Beck et al., 2012; Schuck et al., 2012; Xiong et al., 2010; Karion et al., 2013), there are only few studies so far, which evaluate the regional variability close to the ground (Lowry et al., 2001; Phillips et al., 2013; Townsend-Small et al., 2012; Jackson et al., 2014). Phillips et al. (2013) and Jackson et al. (2014) found methane hotspots (up to 28.6 ppm) along the roads of Boston and up to 54 ppm across Washington DC which were attributed to leakages in the gas distribution network in both studies. Methane concentrations also varied substantially in the greater area of Los Angeles (Townsend-Small et al., 2012). Still, the temporal fluctuations of the spatial variability and the underlying driving processes remained unresolved.

In addition, emission budgets of atmospheric methane are mainly based on modeling approaches and rarely include direct measurements. Currently, two modeling approaches, i.e., top-down and bottom-up, are used to assess methane fluxes (Nisbet and Weiss, 2010). In top-down or backward modeling, often implemented for global methane budgets, atmospheric methane concentrations are traced back to regional sources using transport models (Bousquet et al., 2006). Uncertainties in those models arise from assumptions regarding the methane emissions, the use of atmospheric transport models (Gerbig et al., 2008; Lin and Gerbig, 2005; Tarasova et al., 2010), and the lack of direct measurements for validation. Although a reliable quantification of regional-scale

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methane emissions using top-down models is not yet possible, global flux patterns are relatively well constrained (Bergamaschi et al., 2010; Dlugokencky et al., 2011). For regional-scale estimates, a dense measurement network would be required (Tarasova et al., 2010; Villani et al., 2010), which does currently not exist.

In contrast, bottom-up models estimate methane emissions via up-scaling of emission factors or small-scale flux calculations using statistical land-use and livestock information. However, the results from bottom-up models are still associated with considerable uncertainties (Gerbig et al., 2009; Jagovkina et al., 2000), which originate from the high variability in reported emission factors (Bergamaschi et al., 2010) and the questionable representativeness of small-scale flux calculations for regional scales. Bottom-up emission estimates need to be verified which can be done using spacially highly resolved measurements (Brondfield et al., 2012) or a network of measurement stations in combination with Lagrangian dispersion models (McKain et al., 2012). Thus, direct atmospheric measurements at regional scales and a better understanding how the heterogeneous patterns in atmospheric methane concentrations arise would help to reliably validate or constrain bottom-up and top-down emission estimates and to reduce associated uncertainties.

Our study aims to improve the understanding of spatio-temporal methane concentration patterns using highly resolved, mobile measurements in an agriculturally dominated area in Switzerland. The objectives of our study are (1) to identify the spatio-temporal methane patterns, (2) to investigate the processes controlling such patterns of methane concentrations in complex terrain, and (3) to assess the potential of methane concentration measurements to constrain regional scale emission estimates.

2. Materials and methods

2.1. Site description and transect patterns

We used a mobile setup for methane concentration measurements in the Reuss Valley, situated in central Switzerland. The Reuss Valley is a valley aligned in a northerly direction, with an altitudinal difference of approximately 440 m between the surrounding hilltops and the valley bottom. Agriculture is the dominant land-use type, with 56.5% of the total area (FSO, 2012). Prevailing diurnal wind patterns within the valley are dominated by two wind systems, the along-valley wind system and the slope wind system, resulting in up-valley and upslope winds during daytime and down-valley and downslope winds during nighttime. The concentration measurements were performed during three consecutive days and nights under fair weather conditions in July 2012, covering two predefined transects, the valley transect and the mountain transect (Fig. 1).

The valley transect measurements were performed eight times (transects t1–t8) during 24th–25th of July 2012 and included the Reuss Valley bottom and the surrounding hillsides, covering altitudes between 380 and 716 m above sea level (a.s.l.). During 26th of July, additional measurements (transects t9–t12) were carried out along the mountain transect, including the Zugerberg mountain ridge (1077 m a.s.l.) located east of Lake Zug, to extend the vertical profiles (Table 1).

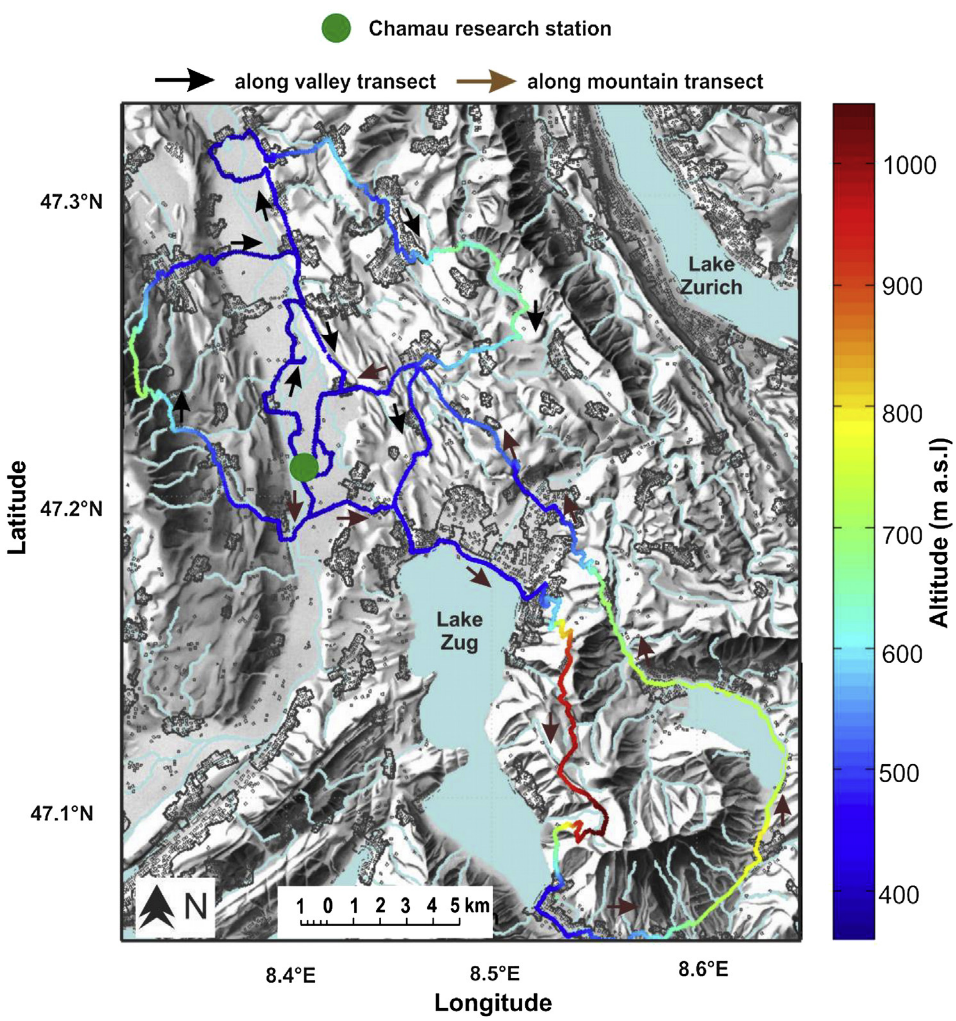

Fig. 1. Routing of the valley (along the black arrows) and the mountain (along the brown arrows) transects overlaid on a relief map of the measurement area including the altitude information at each measurement point (color axis). The relief map was provided by the Federal Office of Topography swisstopo (Art. 30 GeoIV): 5704 000 000. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)