Gap-filling of VOC flux data for deriving annual budgets: A mountain meadow case study

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Introduction

According to the present scientific understanding biogenic volatile organic compounds (BVOCs) influence tropospheric ozone levels [1] and contribute to the formation of secondary organic aerosols [2] and thus play a key role in atmospheric chemistry.
Although the biosphere is currently thought to be the main source for VOCs longer measurements reflecting seasonal or even inter-annual changes of biogenic VOC fluxes barely exist and usually require a filling of data gaps for complete annual information. The lack of long-term measurement data for a comparison with VOC models, which generally work on annual timescales, limits the model accuracy and hinders a more exact VOC quantification.
We execute a systematic comparison of different options for filling gaps in long-term VOC data and provide complete annual time series for the most important BVOCs above grassland.

Material and Methods

Flux measurements of several VOCs were performed at a meadow which is located in the middle of a flat valley bottom close to the village Neustift (47°07' N, 11°19' E) in Stubai valley, Austria at an altitude of 970 m (a.s.l.).

VOC flux data are available for two complete growing seasons of the grassland (from snowfall to snowfall) in the years 2009 and 2011. We use this data series to test the performance of four different gap filling methods on the cumulative fluxes and to retrieve an annual VOC balance for the two years.

Average diurnal cycles (ADIV)
Gap filling on a half-hourly time scale building average diurnal cycles within a specified time window of ± 8 days around the gap.

Daily averages (AGW)
Gap filling on a daily time scale averaging VOC fluxes within a time window of ± 8 days around the missing day.

Look up tables (LUT)
Gap filling defining classes of flux conditions for every day (temperature, PAR, GAI, precipitation) and replacing missing days by class averages.

Linear interpolation (LIP)
Gap filling on a daily time scale using simple linear interpolation

Results, Discussion & Conclusions

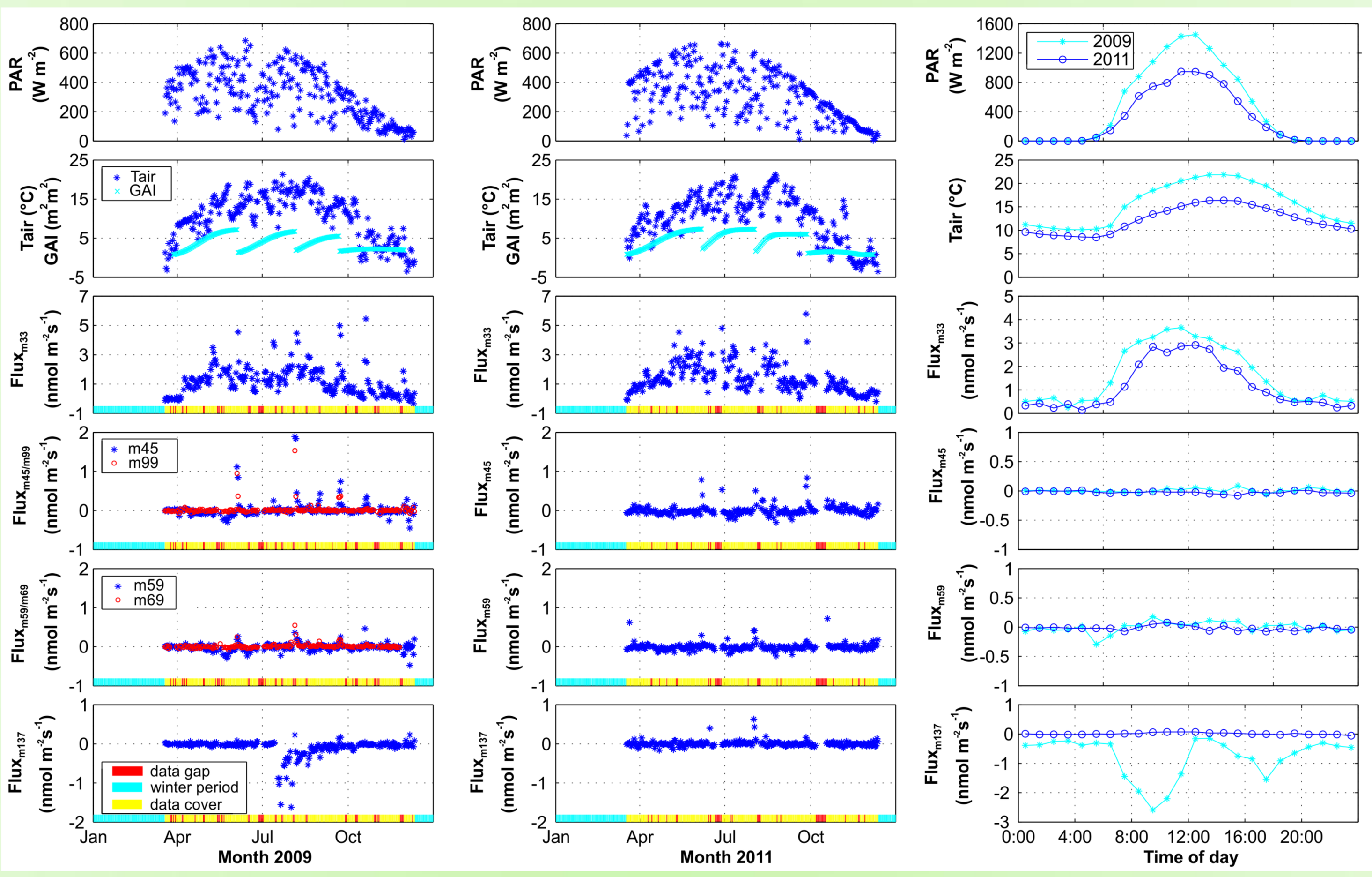

Figure 1: *Measured VOC time series for 2009 and 2011 and diurnal cycles during a selected summer period for every VOC and both years*

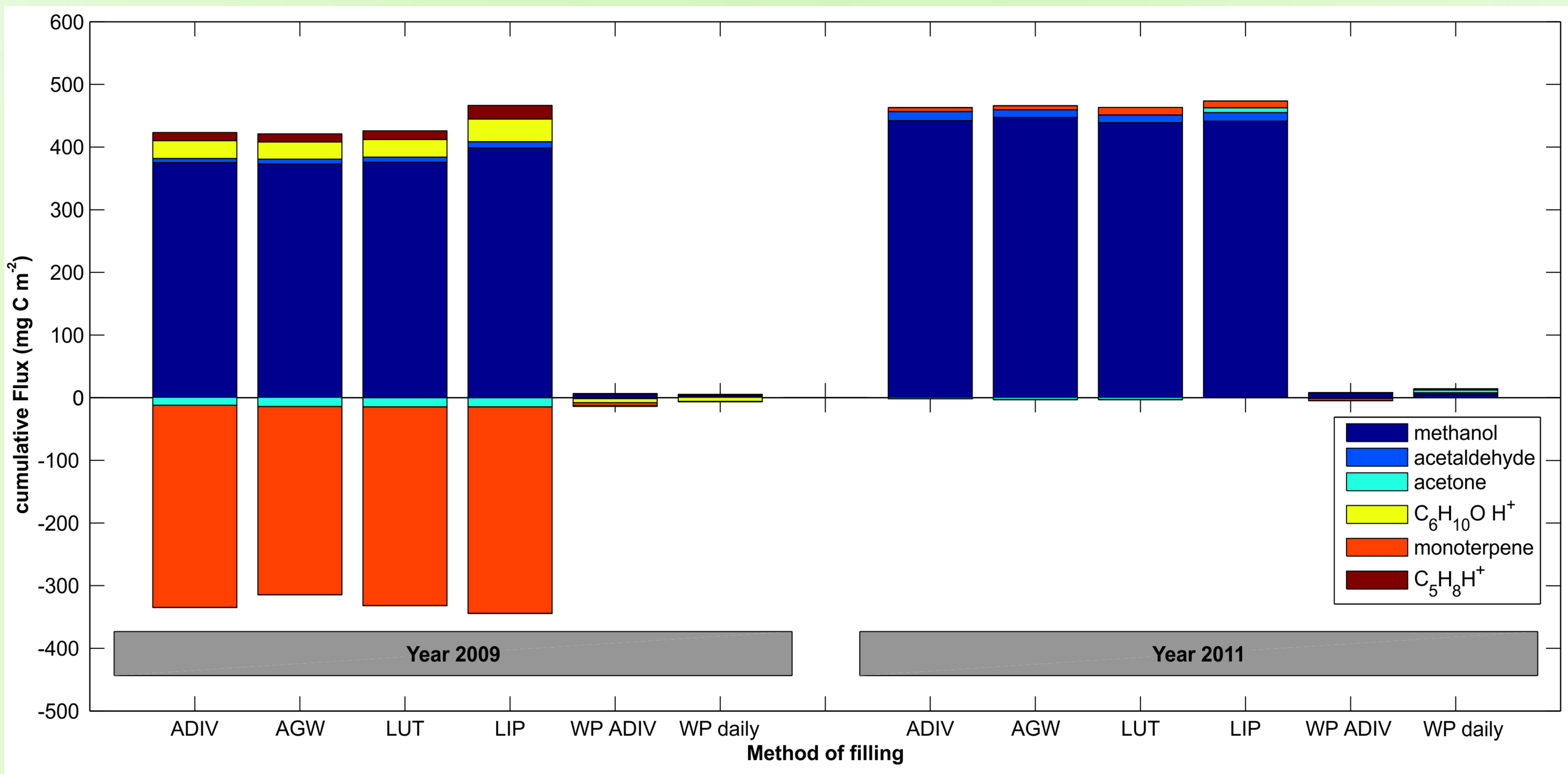

Figure 2: *Comparison of gap-filled cumulative VOC fluxes using the different filling methods for the growing season and the winter period (WP) in 2009 and 2011*

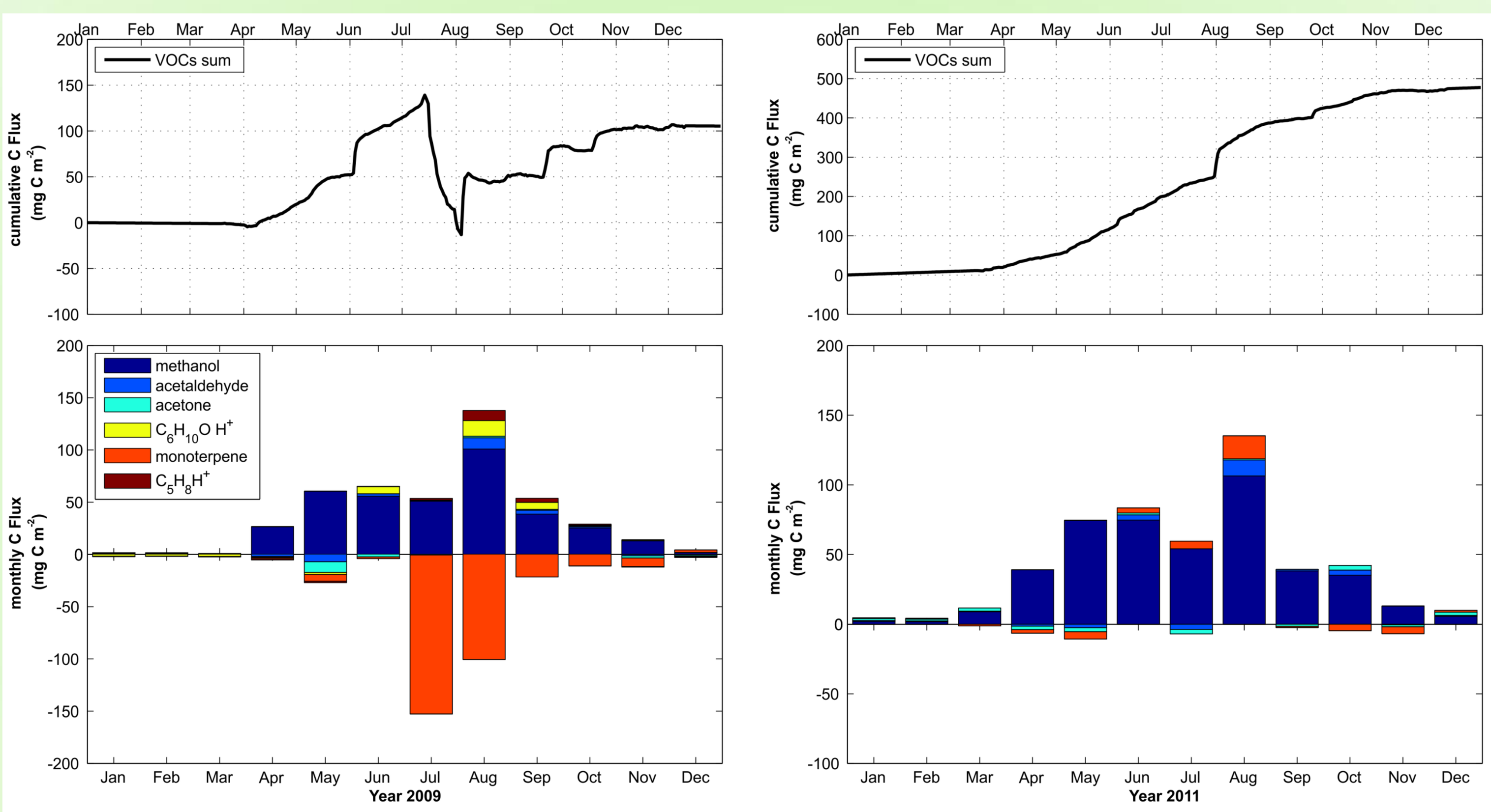

Figure 3: *Cumulative fluxes of the sum of VOCs (upper panels) and monthly VOC fluxes (lower panels).*

Error gap filling			m/z 33	m/z 45	m/z 59	m/z 99	m/z 137	m/z 69	VOC
2009	cummulative flux (mg C m^{-2})		**380.7**	**8.0**	**-13.9**	**30.1**	**-317.4**	**15.3**	**102.8**
	mean error (mg C m^{-2})	ADIV	3.1	0.9	1.0	0.8	13.4	1.1	20.3
		LUT	3.1	1.1	1.3	0.8	21.0	1.1	28.4
		AGW	3.7	1.0	1.2	0.8	18.9	1.3	26.9
		LIP	4.0	1.6	1.7	1.2	26.6	1.8	36.9
2011	cummulative flux (mg C m^{-2})		**442.5**	**13.2**	**-0.1**	**---**	**8.9**	**---**	**464.5**
	mean error (mg C m^{-2})	ADIV	6.6	1.9	1.5	---	3.0	---	13.0
		LUT	7.3	1.4	1.3	---	2.5	---	12.5
		AGW	8.3	1.9	2.2	---	3.6	---	16.0
		LIP	11.7	2.9	3.3	---	6.7	---	24.6

Table 1: *Mean error introduced due to the gap filling for different VOCs*

The gap filling on a half-hourly basis introduced lowest errors to the data but the performance of AGW and LUT method was reasonably good as well. Gap filling by linear interpolation, however, led to considerably higher uncertainties.
The years 2009 and 2011 showed different flux patterns mainly due to considerable deposition fluxes of monoterpenes (-317 mg C m-2) in the aftermath of a hailstorm in 2009. Methanol shows, with emission values of 381 mg C m^{-2} (2009) and 443 mg C m^{-2} (2011), highest fluxes in both years. Other compounds exhibited considerably lower fluxes.

[1] R. Atkinson, Atmospheric chemistry of VOCs and Nox, Atmospheric Environment, 34, 2063-2101, 2000
[2] M. Hallquist et al., The formation, properties and impact of secondary organic aerosol: current and emerging issues, *Atmospheric Chemistry and Physics*, 9, 5155-5236, 2009