

S1 Point Sources PCH4 emissions inventory

see the attached ascii file for details on the coal mines and single shafts, their exact locations, methane emissions and the corresponding references.

S2 Flight pattern for P4 and P5

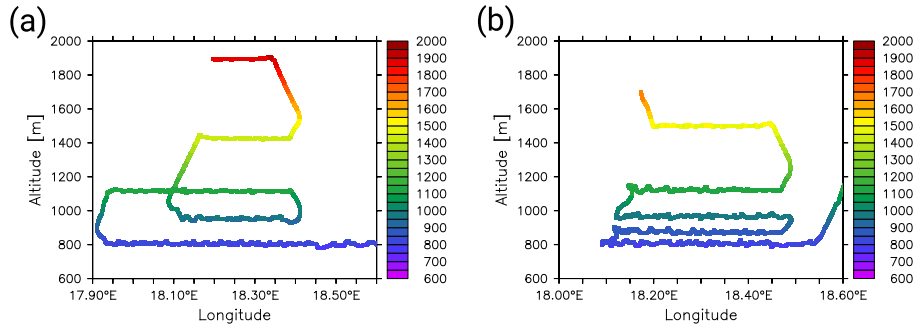


Figure S1: Panel (a) shows the flight pattern for P4, panel (b) shows the flight pattern for P5. Colors in the key refer to the altitude.

S3 Comparisons of P1, P3, P6 and P7 to the simulated CH4.FX of CM7 and CM2.8

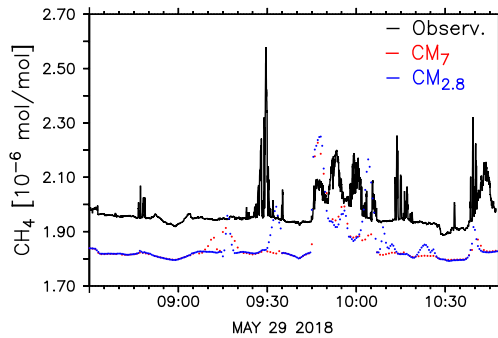


Fig. S2: Methane mixing ratios of P1 on May 29, 2018. Observations are in black, model results of CM7 and CM2.8 are in red and blue, respectively.

At 09:30 UTC D-FDLR flew close to the ventilation shafts resulting in high observed methane mixing ratios. This peak is only resolved by CM2.8, even though it is shifted in time or in space. The model performance decreases if the measurements are taken very close to the ventilation shafts. This is also seen for P2 and P3, where localized methane enhancements cannot be resolved by the model. Between 09:45 and 10:10 UTC, the observed methane enhancements are

simulated in both model instances, but show very high amplitudes compared to the observations.

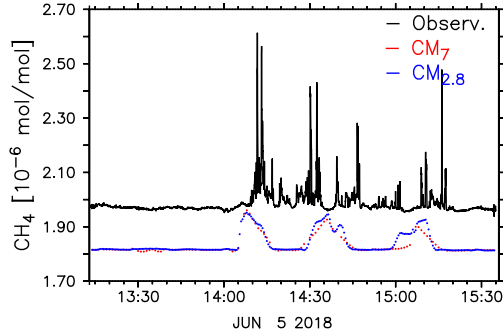


Fig. S3: Methane mixing ratios of P3 on June 05, 2018. Observations are in black, model results of CM7 and CM2.8 are in red and blue, respectively.

The comparison between D-FDLR in-situ observations and the model show that the observed peaks can be simulated, but observations show more variability and the simulated methane peaks are shifted in time or in space, which results in a low correlation in the Taylor diagram (see Fig. 11). When catching the methane plume, the aircraft flew very close to the ventilation shafts, which resulted in high mixing ratios up to $2.65 \cdot 10^{-6}$ mol/mol. Although in general, simulated peak amplitudes and observed peak amplitudes are in good agreement, the model is not able to resolve these short-term enhancements (see also Figure S2).

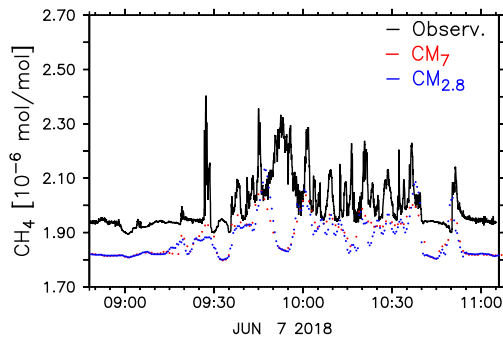


Fig. S4: Methane mixing ratios of P5 on June 07, 2018. Observations are in black, model results of CM7 and CM2.8 are in red and blue, respectively.

On June 07, model results agree well with the observed methane peaks. Besides the enhancement at 09:30 UTC, amplitudes are very similar to those of the observations. This can be also seen in the Taylor diagram (see Fig. 11),

where P6 is close to the red reference line. Between 09:50 UTC and 10:00 UTC, D-FDLR flew close to the beginning of the Sudetes, right after the Czech boarder. Next to increased methane mixing ratios, they also observed high mixing ratios of CO and CO₂ here. These gases might have accumulated north east of the mountain range but the model is not able to simulate this feature.

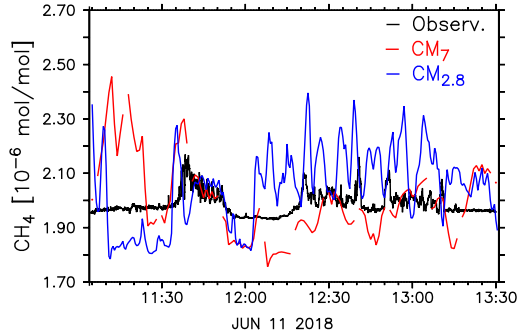


Fig. S5: Methane mixing ratios of P7 on June 11, 2018. Observations are in black, model results of CM7 and CM2.8 are in red and blue, respectively.

On June 11 wind blew predominantly from north west and the observed peak around 11:40 UTC can be attributed to a the southwestern USCB mines. After 12:20 UTC, D-FDLR flew downwind of the norther USCB mines. Both model instances are able to simulate the first methane peak around 11:40 UTC, but differ from the observations before and after that peak. The model shows very high amplitudes and high variability in methane mixing ratios. High wind speeds might be the reason. The correlation coefficients of both instances is below 0.3 and the normalized standard deviation is 3.5 and 3.6 for CM2.8 and CM12, respectively. Consequently P7 lies outside the Taylor diagram in Fig.11.