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An improved method for mobile characterisation of $\delta^{13}CH_4$ source signatures and its application in Germany

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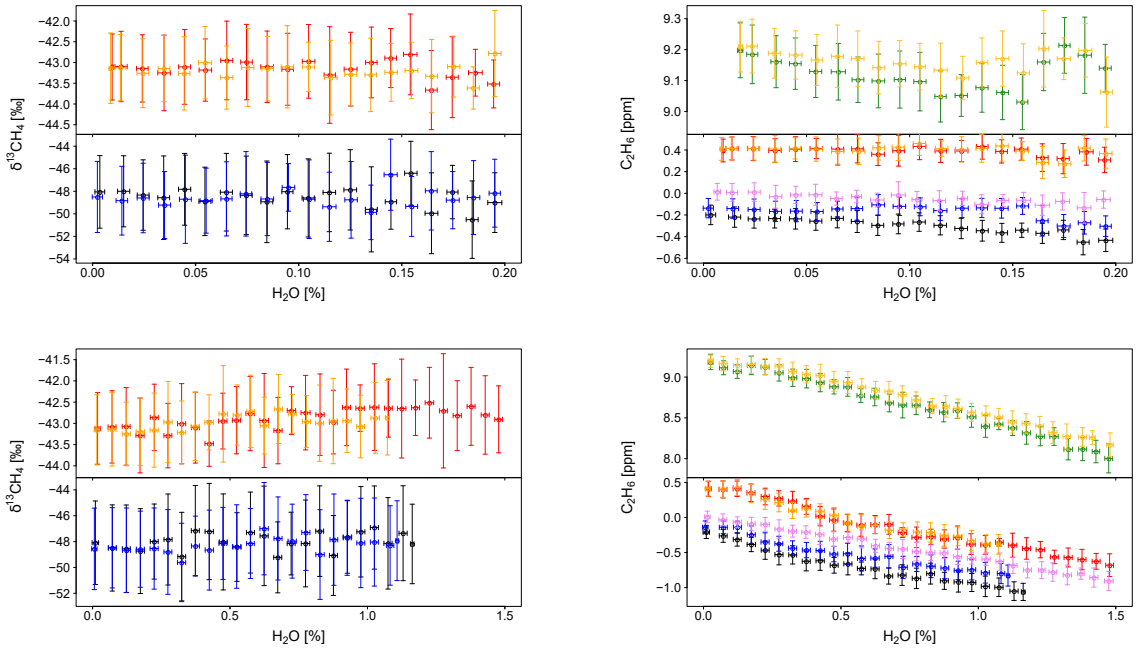

Figure S1. H_2O interference on $\delta^{13}CH_4$ and C_2H_6. Different colours indicate different tests and in part different gas cylinders used.

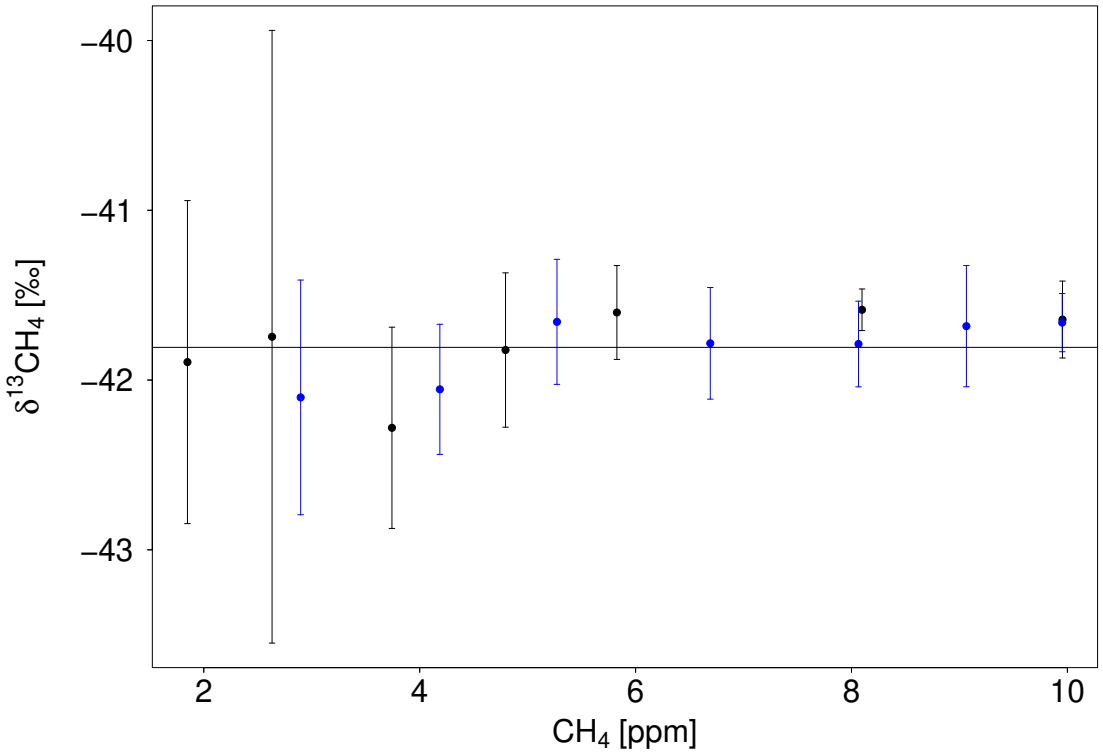

Figure S2. CH_4 interference on $\delta^{13}CH_4$. The points show the 15 min average measurement of different gas mixtures prepared with natural air (N_2, O_2 and Ar) and dry compressed air of 10 ppm CH_4 (in N_2, O_2 and Ar). The same dilution test was done twice (blue and black points).

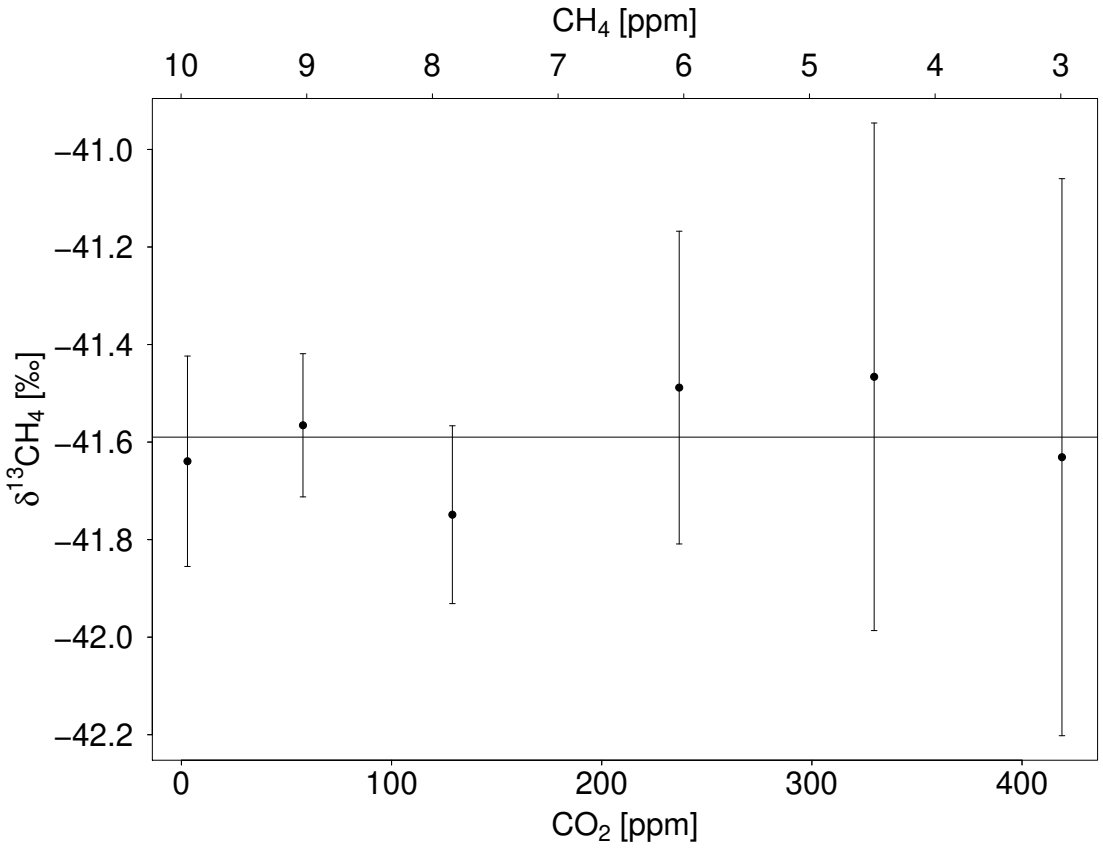

Figure S3. CO_2 interference on $\delta^{13}CH_4$. The points show the 15 min average measurement of different gas mixtures prepared by a dilution test with dry compressed air of 10 ppm CH_4 (in N_2, O_2 and Ar) and dry compressed air of 600 ppm CO_2 (in N_2, O_2 and Ar).

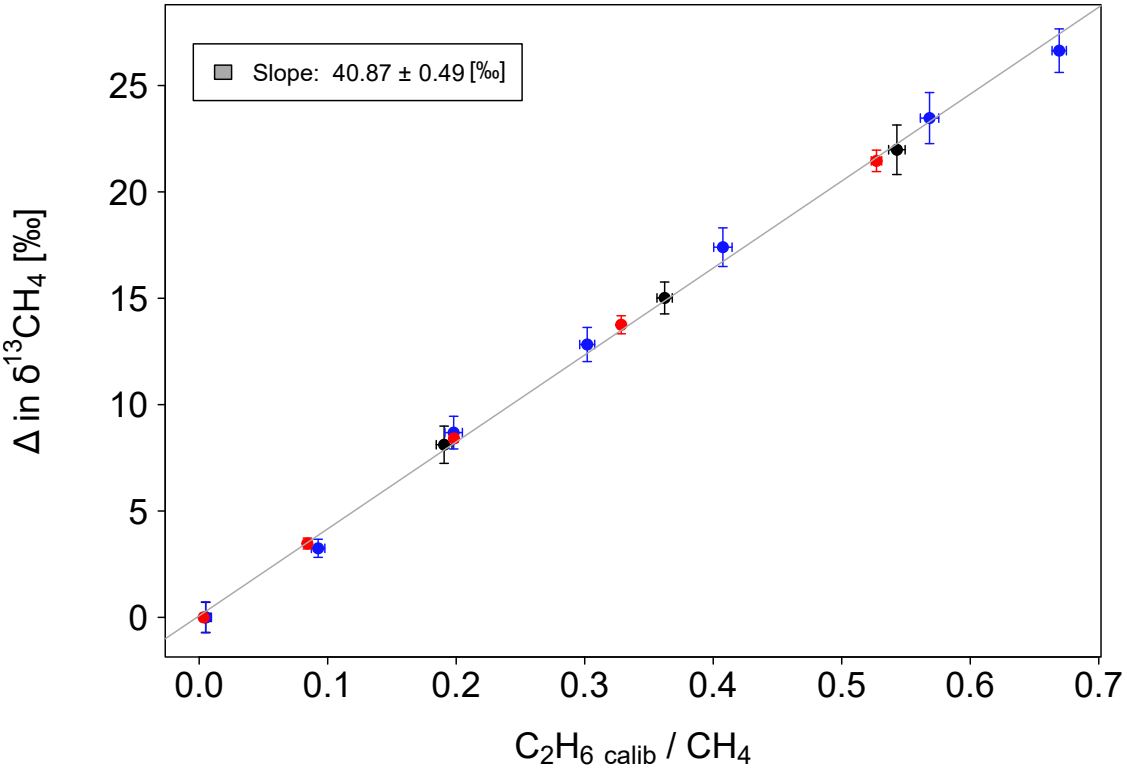

Figure S4. C_2H_6 interference on $\delta^{13}CH_4$. The points show the corrected and calibrated 15 min average of different gas mixtures prepared by three dilution test with dry compressed air of 5 ppm C_2H_6 (in N_2, O_2 and Ar) and dry compressed air of 10 ppm CH_4 (in N_2, O_2 and Ar, red) or atmospheric concentrations (black and blue).

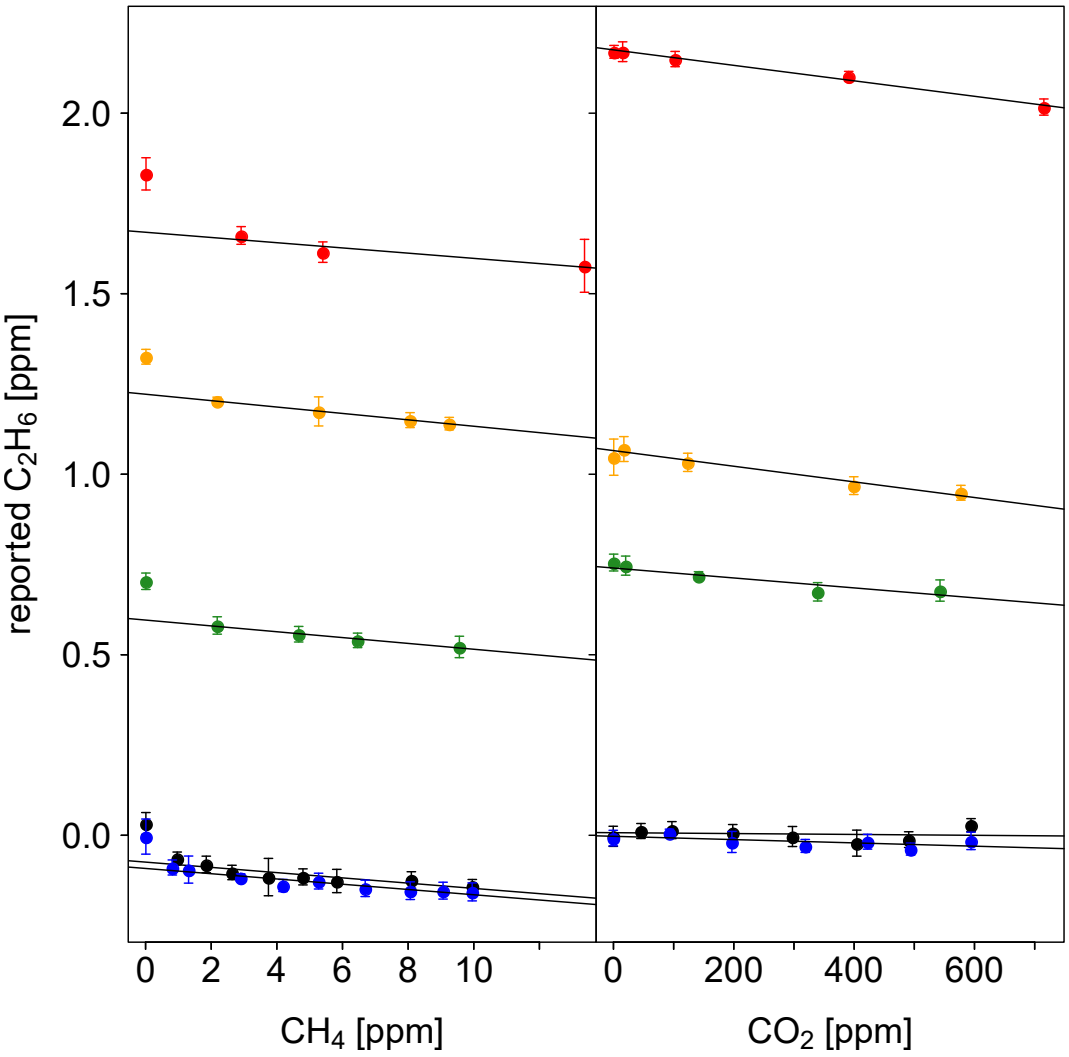

Figure S5. CH_4 and CO_2 interference on C_2H_6. The points show the reported 10−15 min average of different gas mixture. The blue and black points show four dilution test with natural air (N_2, O_2 and Ar) and dry compressed air of 10 ppm CH_4 (in N_2, O_2 and Ar, red) or 600 ppm CO_2 (in N_2, O_2 and Ar). The red, yellow and green measurements show injection tests at which different amounts of pure CH_4 or CO_2 were injected into three liter sample bags filled with natural air (N_2, O_2 and Ar) and different amounts of C_2H_6.

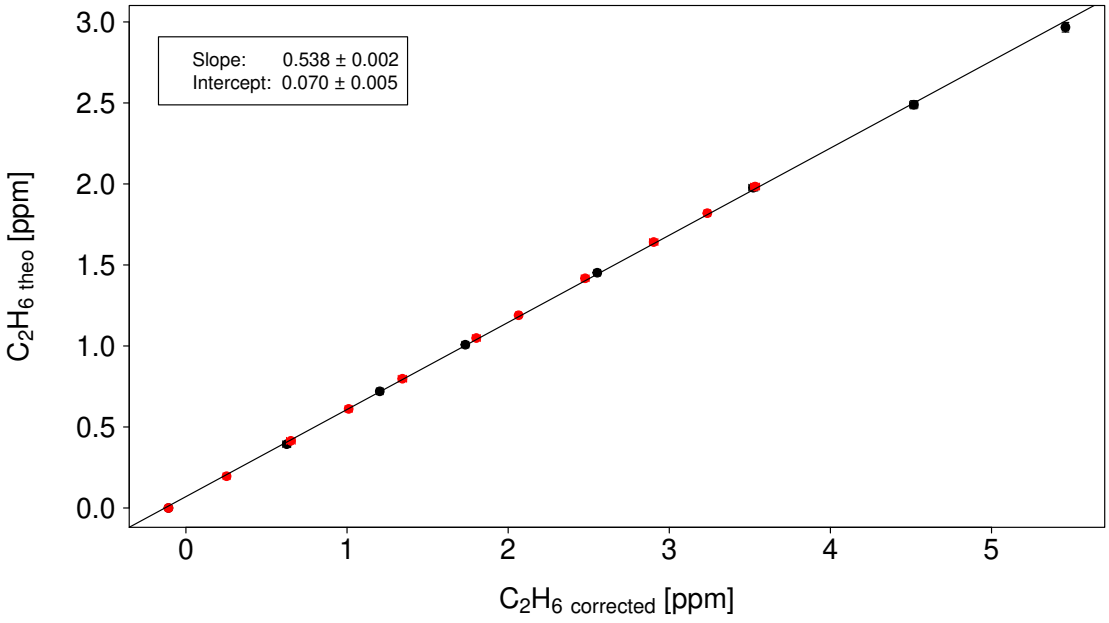

Figure S6. Linearity test of C_2H_6. The points (black and red) show the corrected $10-15$ min average of different gas mixture prepared by two dilution tests with dry compressed air of atmospheric concentrations and dry compressed air of 5 ppm C_2H_6 (in N_2, O_2 and Ar). The theoretical C_2H_6 concentrations were calculated using the measured CH_4 and CO_2 concentrations in the gas mixture and the known CO_2, CH_4 and C_2H_6 concentrations of the used gas cylinders.

Table S1. $\delta^{13}CH_4$ signatures determined for each AirCore measurement.

location	date	$\delta^{13}CH_4$ [‰]	r^2	peak height above baseline
biogas plant				
Heidelberg	2016-08-29	-62.69 ± 1.05	0.939	2.59
	2016-09-08	-62.04 ± 0.20	0.999	9.45
	2016-09-08	-61.23 ± 0.60	0.994	4.85
	2016-09-28	-58.96 ± 0.57	0.992	8.15
	2016-09-28	-61.89 ± 0.32	0.997	7.39
	2016-09-28	-59.81 ± 1.03	0.959	2.34
	2016-10-10	-64.16 ± 0.44	0.996	7.34
	2016-10-10	-63.66 ± 1.06	0.992	3.77
	2016-11-30	-63.15 ± 0.58	0.995	6.70
	2016-11-30	-63.11 ± 0.97	0.993	3.76
	2016-11-30	-63.46 ± 0.74	0.993	4.44
	2016-12-19	-61.99 ± 3.25	0.932	1.27
	2016-12-19	-62.11 ± 1.33	0.984	2.55
	2017-02-22	-67.43 ± 1.26	0.995	4.02
	2017-02-22	-63.58 ± 0.84	0.987	3.64
	2017-02-22	-60.85 ± 0.95	0.981	2.85
	2017-02-22	-61.12 ± 1.63	0.965	3.55
dairy farm				
Weinheim (on farm)	2016-10-26	-64.92 ± 0.71	0.994	6.32
	2016-10-26	-62.62 ± 0.52	0.996	6.67
	2016-11-21	-65.99 ± 0.98	0.950	5.28
Weinheim (plume with biogas plant)	2016-09-29	-62.58 ± 2.08	0.936	1.61
	2016-09-29	-60.16 ± 2.09	0.970	2.09
	2016-09-29	-59.59 ± 1.59	0.965	2.44
	2016-10-26	-56.58 ± 1.20	0.972	2.95
	2016-10-26	-59.66 ± 0.14	0.999	10.91
	2016-11-21	-60.43 ± 0.85	0.990	4.08
	2016-12-14	-47.17 ± 0.25	0.998	10.83
	2016-12-14	-43.13 ± 1.15	0.986	4.19
	2017-02-23	-47.18 ± 2.82	0.950	1.90
	2017-02-23	-43.64 ± 1.55	0.988	3.57
Ladenburg (on farm)	2016-10-26	-64.00 ± 2.63	0.981	1.97
	2016-10-26	-61.56 ± 2.51	0.955	1.56
	2016-10-26	-63.93 ± 0.88	0.990	4.69
Ladenburg (plume with biogas plant)	2016-11-30	-40.30 ± 0.42	0.986	5.95
	2016-11-30	-40.36 ± 0.59	0.974	5.45
	2016-11-30	-41.77 ± 0.93	0.968	5.09
	2016-11-30	-55.08 ± 1.48	0.718	1.74
Kleve (organic farming)	2017-03-24	-63.77 ± 0.42	0.997	8.45
	2017-03-24	-65.11 ± 1.75	0.979	2.86
Kleve (conventional farming)	2017-03-24	-65.37 ± 0.42	0.998	8.04
	2017-03-24	-63.30 ± 0.18	0.998	11.20
Kleve (plume)	2017-03-24	-61.65 ± 1.67	0.976	2.70

Table S1. $\delta^{13}CH_4$ signatures determined for each AirCore measurement.

location	date	$\delta^{13}CH_4$ [‰]	r^2	peak height above baseline
landfill				
Sinsheim (plume)	2016-08-29	-54.16 ± 4.45	0.470	0.47
	2016-09-08	-62.19 ± 4.36	0.582	0.54
	2016-11-02	-59.56 ± 4.99	0.498	0.46
	2016-11-30	-58.96 ± 4.63	0.523	0.50
Sinsheim (on landfill)	2017-07-18	-59.12 ± 1.14	0.992	4.86
	2017-07-18	-59.87 ± 2.80	0.947	1.77
	2016-07-25	-64.86 ± 0.97	0.987	3.42
	2016-07-25	-67.68 ± 2.86	0.914	0.93
	2016-07-25	-69.30 ± 2.99	0.850	0.94
	2016-07-25	-63.99 ± 4.64	0.606	0.47
WWTP				
Heidelberg	2016-10-10	-50.80 ± 2.28	0.935	1.65
	2016-10-26	-51.17 ± 3.97	0.823	1.29
	2016-10-26	-56.34 ± 1.20	0.988	3.85
	2016-12-14	-51.97 ± 1.41	0.977	3.28
	2016-12-19	-54.21 ± 2.31	0.960	1.75
	2017-02-22	-54.22 ± 2.41	0.930	2.04
	2017-02-22	-49.40 ± 2.78	0.837	1.63
natural gas facilities				
Sandhausen	2016-11-02	-42.14 ± 0.59	0.994	7.58
	2016-11-02	-41.50 ± 1.00	0.967	3.05
	2017-03-09	-49.16 ± 4.58	0.810	1.00
Hähnlein/Gernsheim	2016-09-29	-41.14 ± 1.40	0.980	2.87
	2016-10-26	-57.45 ± 2.69	0.922	1.31
	2016-11-21	-51.04 ± 4.00	0.841	1.20
	2016-11-21	-47.60 ± 3.67	0.938	1.63
	2016-11-21	-44.49 ± 1.69	0.957	2.86
	2016-11-21	-47.35 ± 2.95	0.894	1.74
	2016-11-21	-51.00 ± 1.03	0.994	5.49
	2016-12-14	-45.11 ± 1.70	0.973	2.97
	2017-02-23	-41.11 ± 3.08	0.851	1.38
bituminous deep coal mine				
Bottrop (active)	2017-03-25	-59.53 ± 2.18	0.947	1.35
	2017-03-25	-54.73 ± 2.28	0.928	1.46
	2017-03-25	-55.04 ± 1.25	0.972	2.29
	2017-03-25	-54.86 ± 0.51	0.996	5.52
Bottrop (closed)	2017-03-25	-49.97 ± 6.33	0.677	0.55