S1 Simulation levels in ARTM

In ARTM the vertical resolution can be customized by the user. If not specified, the default resolution shown in Table S1 is used. For the comparison of simulation results of the Bełchatów power plant with observations a finer vertical grid resolution shown in Table S2 was used.

Table S1. Default setup of the horizontal levels in ARTM. The height of the lower level boarder above ground level (agl) and the level thickness are given in meter.

level	height of lower level boarder [m agl]	thickness [m]
19	1200	300
18	1000	200
17	800	200
16	700	100
15	600	100
14	500	100
13	400	100
12	300	100
11	200	100
10	150	50
9	100	50
8	65	35
7	40	25
6	25	15
5	16	9
4	10	6
3	6	4
2	3	3
1	0	3

Table S2. Horizontal levels used for the simulations when comparing ATRM with observations. The height of the lower level boarder agl and the level thickness are given in meter.

level	height of lower level boarder [m agl]	thickness [m]
41	1800	100
40	1750	50
39	1700	50
38	1650	50
37	1600	50
36	1550	50
35	1400	50
34	1350	50
33	1300	50
32	1250	50
31	1200	50
30	1150	50
29	1100	50
28	1050	50
27	1000	50
26	950	50
25	900	50
24	850	50
23	800	50
22	750	50
21	700	50
20	650	50
19	600	50
18	550	50
17	500	50
16	450	50
15	400	50
14	350	50
13	300	50
12	250	50
11	200	50
10	150	50
9	100	50
8	65	35
7	40	25
6	25	15
5	16	9
4	10	6
3	6	4
2	3	3
1	0	3

S2 Additional time series for well-mixed condition test

The following figures show the temporal evolution of the normalised concentration $c\overline{c}^{-1}$ in certain height levels for a duration of 30 days (720 h). The shown levels are at 12.5 m, 337.5 m and 1087.5 m height for the different turbulence models ARTM2, ARTM3, PRFMOD, MODHANNA and DEGRAZIA. The time axes are split into two different scales.

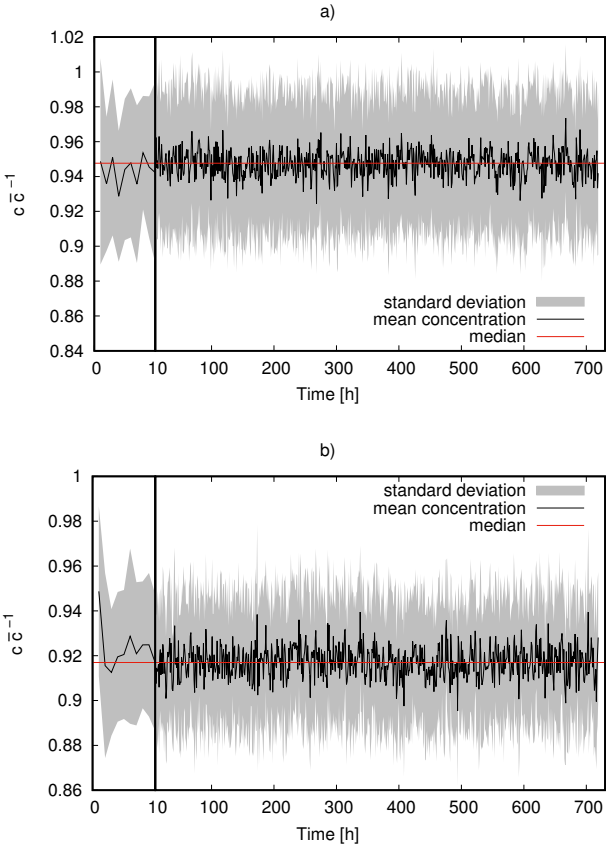

Figure S1. Time series of the normalised concentration for the ARTM2 turbulence model at a) $z\,h_{\mathrm{m}}^{-1} \approx 0$ (12.5 m height) and b) $z\,h_{\mathrm{m}}^{-1} \approx 1$ (1087.5 m height) for 30 days (720 h).

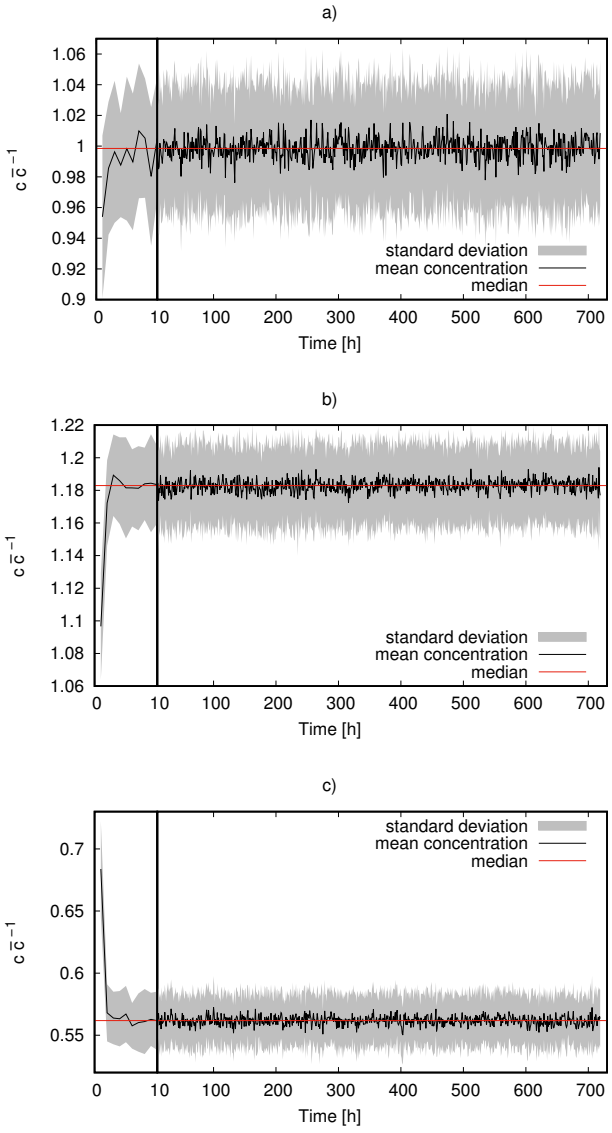

Figure S2. Time series of the normalised concentration for the ARTM3 turbulence model at a) $z\,h_{\mathrm{m}}^{-1} \approx 0$ ($12.5\,\mathrm{m}$ height), b) $z\,h_{\mathrm{m}}^{-1} \approx 0.3$ ($337.5\,\mathrm{m}$ height) and c) $z\,h_{\mathrm{m}}^{-1} \approx 1$ ($1087.5\,\mathrm{m}$ height) for 30 days ($720\,\mathrm{h}$).

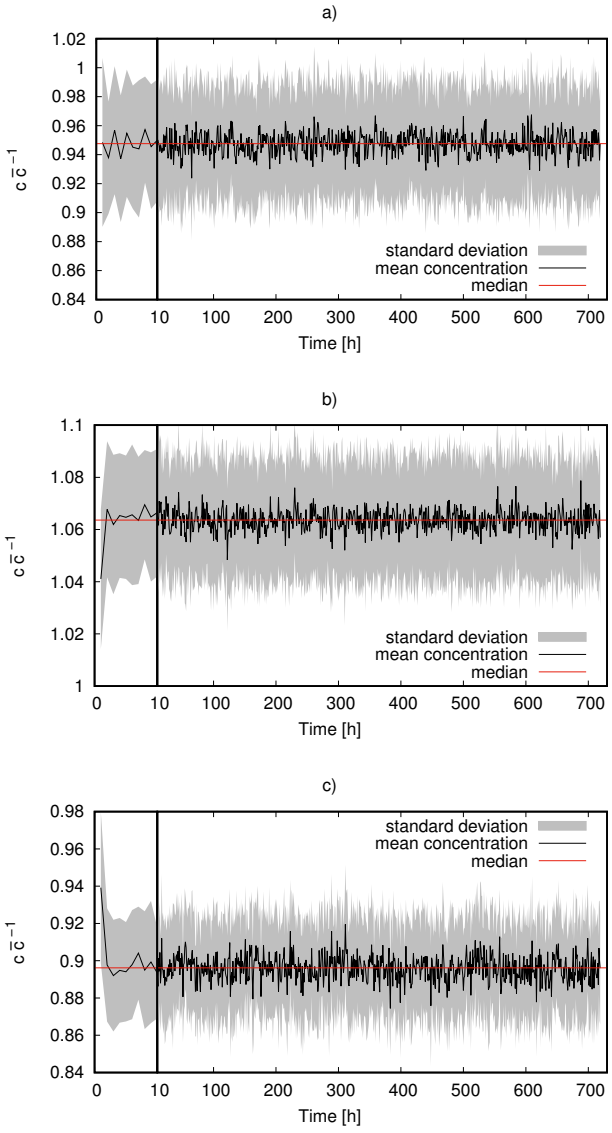

Figure S3. Time series of the normalised concentration for the PRFMOD turbulence model at a) $z\,h_{\mathrm{m}}^{-1} \approx 0$ (12.5 m height), b) $z\,h_{\mathrm{m}}^{-1} \approx 0.3$ (337.5 m height) and c) $z\,h_{\mathrm{m}}^{-1} \approx 1$ (1087.5 m height) for 30 days (720 h).

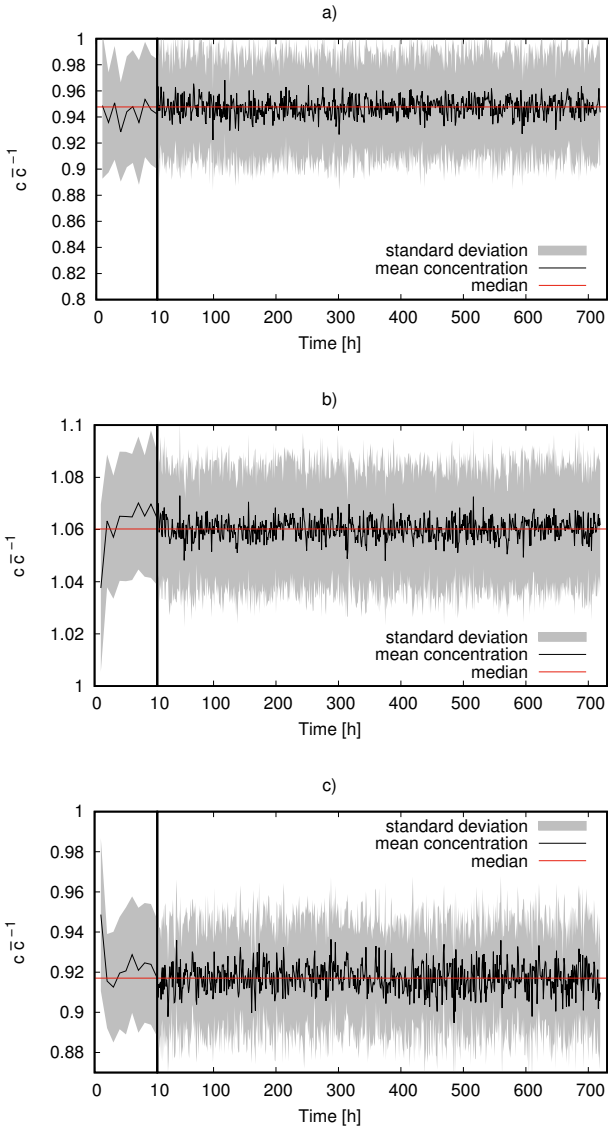

Figure S4. Time series of the normalised concentration for the MODHANNA turbulence model at a) $z\,h_{\mathrm{m}}^{-1} \approx 0$ (12.5 m height), b) $z\,h_{\mathrm{m}}^{-1} \approx 0.3$ (337.5 m height) and c) $z\,h_{\mathrm{m}}^{-1} \approx 1$ (1087.5 m height) for 30 days (720 h).

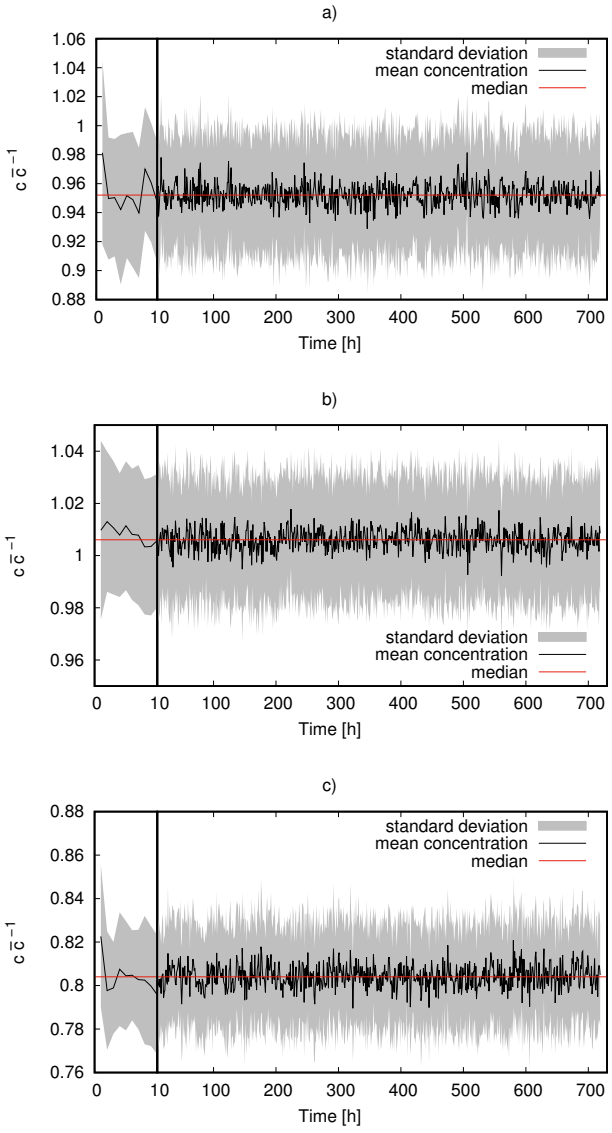

Figure S5. Time series of the normalised concentration for the DEGRAZIA turbulence model at a) $z\,h_{\mathrm{m}}^{-1} \approx 0$ (12.5 m height), b) $z\,h_{\mathrm{m}}^{-1} \approx 0.3$ (337.5 m height) and c) $z\,h_{\mathrm{m}}^{-1} \approx 1$ (1087.5 m height for 30 days (720 h).

S3 Measurement data from the aircraft flight

The collected measurement data from the measurement flight in the vicinity of the Bełchatów power plan is given in Fig. S6. The original data had been transformed to the height above ground level instead of the altitude above mean sea level.

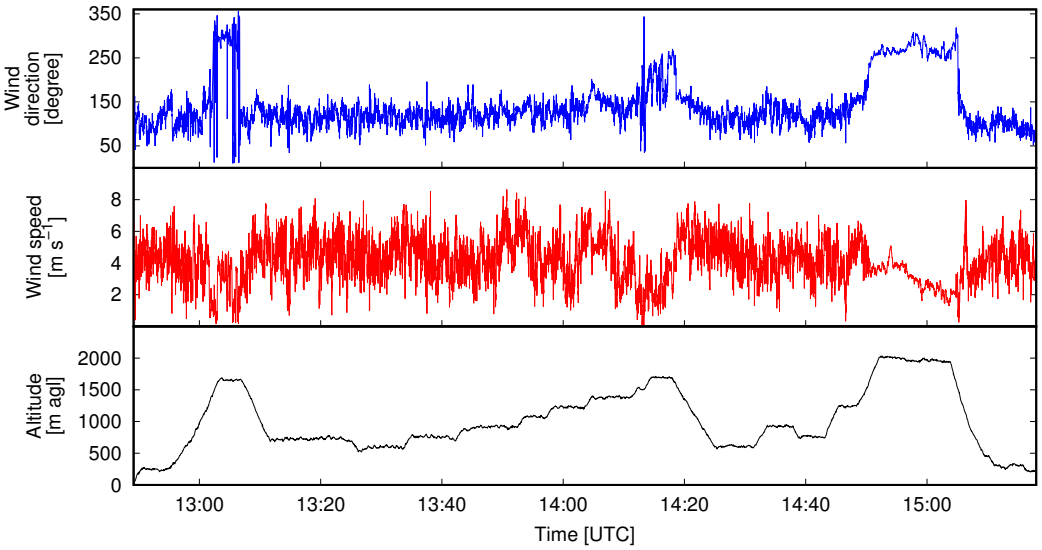

Figure S6. Wind direction, wind speed and flight height measured by the aircraft during the measurement flight.

S4 Description of parameter derivation for the comparison of simulations with observations

The stability class (SC) was determined according to the scheme given in KTA 1508 (2017) from the horizontal wind direction fluctuations measured on several transects at different height levels and was classified as "very unstable" during the observations (Klug, 1969). For the determination of the roughness length z_0, the CORINE Land Cover Inventory of 2018 and the categorisation after TA Luft (2002) was used. The area is covered mainly by arable land, pastures, coniferous and mixed forest leading to a mean value of $z_0 = 0.5\,\mathrm{m}$ for the simulation domain. The zero plane displacement was assumed to be $d_0 = 6 \cdot z_0$ (TA Luft, 2002). The mixing layer height of $1650\,\mathrm{m}$ was derived from the observations by locating the abrupt decrease in the wind speed fluctuation (see Fig. S6). The elevation data originates from the Shuttle Radar Topography Mission version 3 (SRTM3) that has a spatial resolution of 3 arc-seconds ($\approx 90\,\mathrm{m}$) (Farr et al., 2007).

The stacks are assumed to have had different emission rates (two-thirds : one-third) because photographs (see Fig. S2) taken from the aircraft showed markedly different plume rise heights for the two stacks. This plume rise is assumed to be $202\,\mathrm{m}$ and $74\,\mathrm{m}$, respectively.

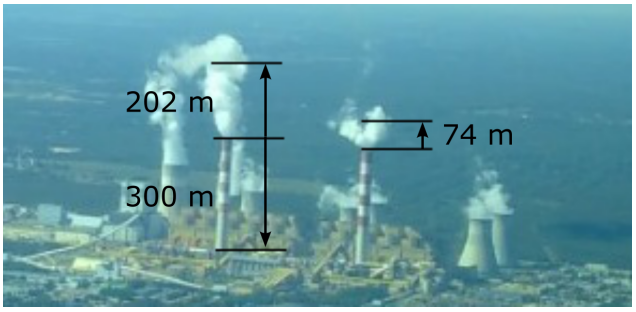

Figure S7. Photograph of the Bełchatów power plant taken by Alina Fiehn from the measuring aircraft during the measurement flight on 7 June 2018 at 13:13 UTC. The stack height as well as the plume rise for both stacks is given in the image. The photograph was taken from the south-south-east of the power plant.

S5 Hourly wind inputs for the simulations to compare with observations

Additional spin-up time before the measurement flight is simulated to ensure a fully developed plume within the simulation domain. For the single wind direction case, the mean wind speed of $4.4\,\mathrm{m\,s^{-1}}$ from the reference transect was used for the simulation time period at approx. $600\,\mathrm{m}$. For the dual wind direction case, reanalysis data from ERA5 were used for the spin-up at the $925\,\mathrm{hPa}$ pressure level ($\approx 600\,\mathrm{m}$). The hourly wind data are shown in Table S3.

Table S3. Hourly inputs for wind direction and wind velocity at $599\,\mathrm{m}$ height for the two cases: one mean wind direction; and two alternating wind directions for the time from 13:00 to 15:00 UTC. The time stamps describe the full hour before the time given in the time column. The measurement flight was performed during step 14:00 and 15:00 UTC and is marked with [1)]. Data from the ECMWF ERA5 data set for $925\,\mathrm{hPa}$ pressure level ($\approx 600\,\mathrm{m}$ agl) are marked with [2)].

Time [UTC]	Single wind direction		Dual wind direction	
	wind direction [degree]	wind velocity [$\mathrm{m\,s^{-1}}$]	wind direction [degree]	wind velocity [$\mathrm{m\,s^{-1}}$]
09:00	120	4.4	101 [2)]	7.1 [2)]
10:00	120	4.4	107 [2)]	6.1 [2)]
11:00	120	4.4	105 [2)]	5.7 [2)]
12:00	120	4.4	103 [2)]	5.6 [2)]
13:00	120	4.4	106	4.4
14:00 [1)]	120	4.4	134	4.4
15:00 [1)]	120	4.4	106	4.4

S6 Additional heat maps for the comparison of simulations and observations

The following Figs. S8 to S10 show the comparison of the simulated and observed plumes in different heights from 550 m to 1100 m agl. The Fig. S11 shows the cross section of the plumes at wall 2.

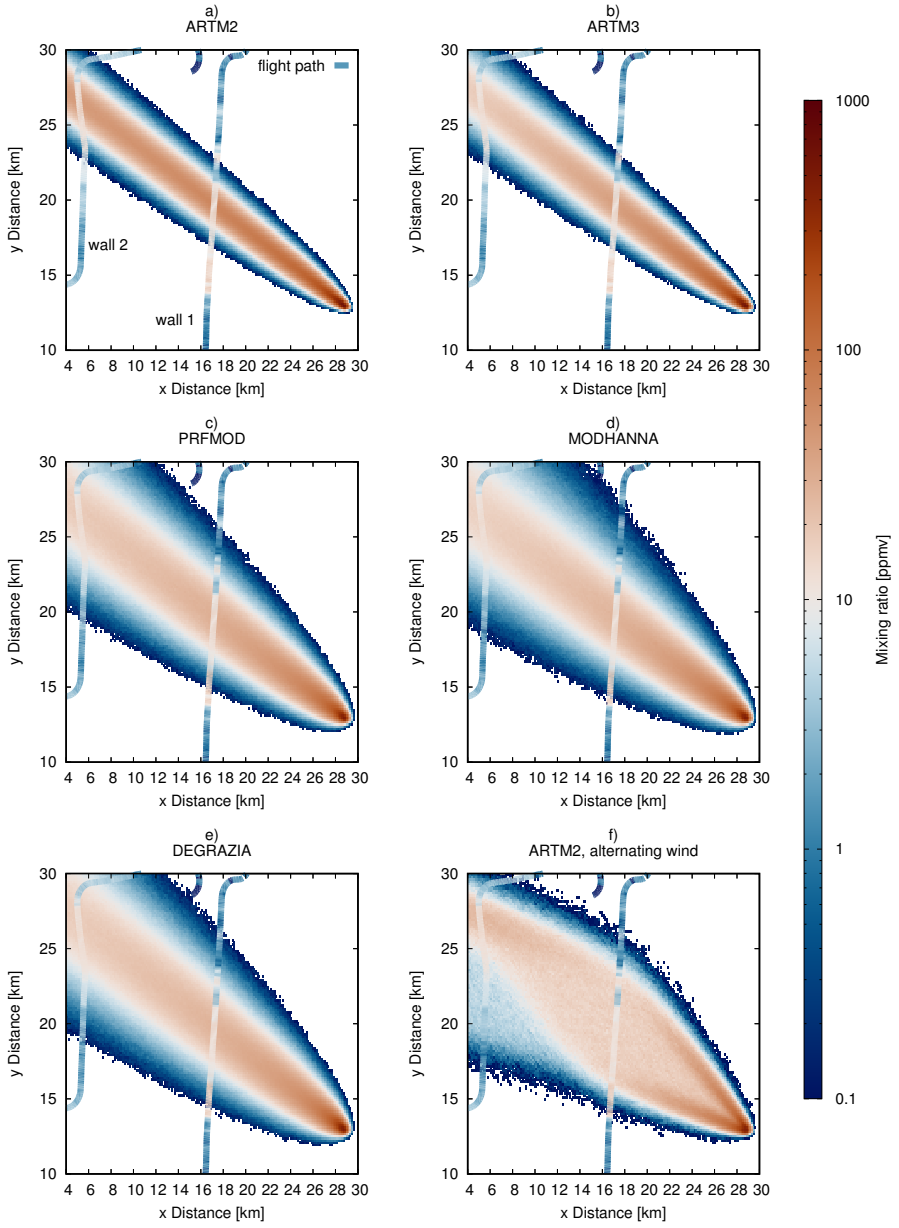

Figure S8. Comparison of the observed (550 m to 650 m agl) and the simulated CO_2 mixing ratio (600 m to 650 m agl).

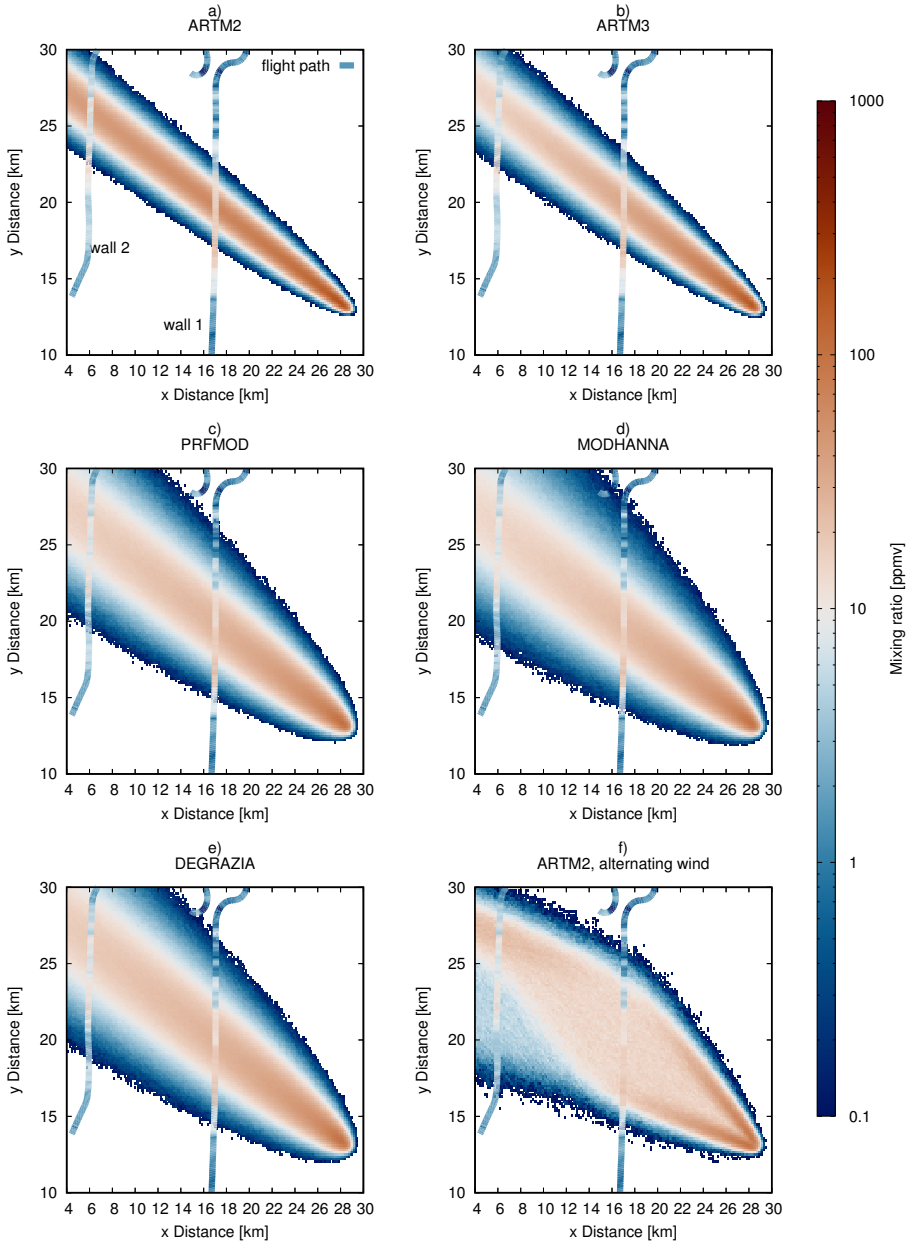

Figure S9. Comparison of the observed (850 m to 950 m agl) and the simulated CO_2 mixing ratio (900 m to 950 m agl).

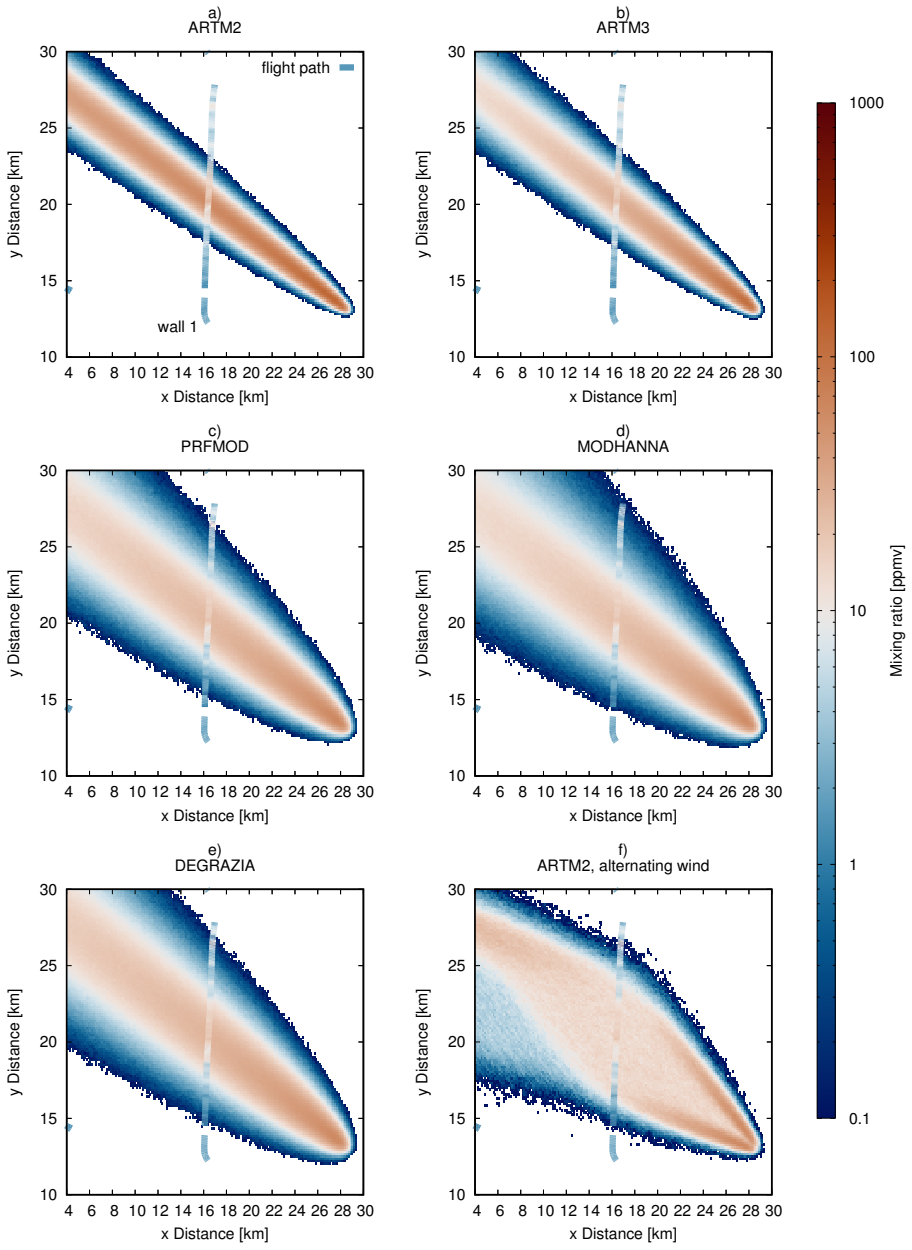

Figure S10. Comparison of the observed and the simulated CO_2 mixing ratio at the height of 1050 m to 1100 m agl.

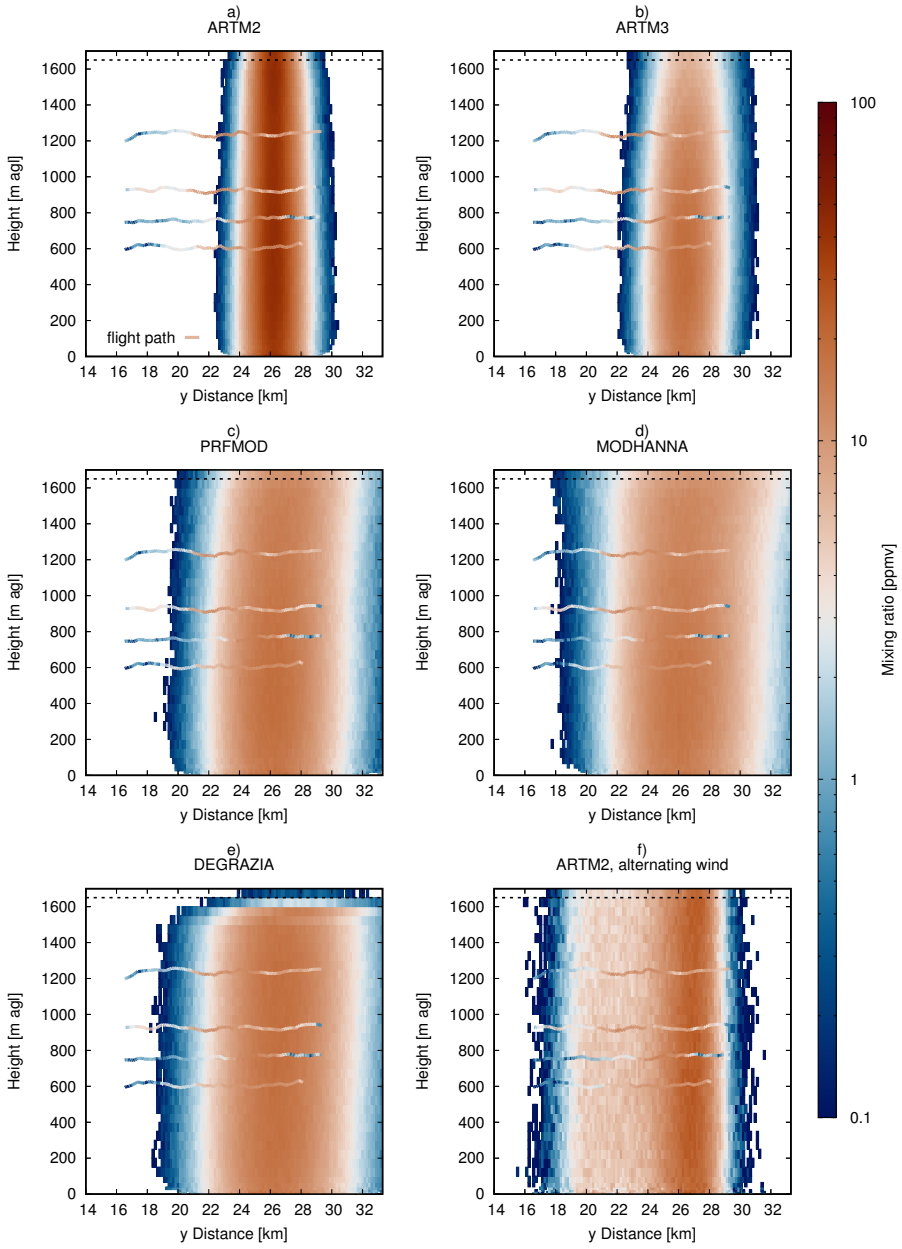

Figure S11. Cross section of wall 2 of the CO_2 plume and the simulated plumes for the different turbulence models. The dotted line is the simulated PBL top. The right boarder of the graphs represent the northern simulation domain boarder.

S7 Description of the limits of the Z statistic

According to the following limits the Z statistic is interpreted as (University of Oregon, 2020):

$Z < 2.0$ two samples are the same,

$2.0 \leq Z < 2.5$ two samples are marginally different,

$2.5 \leq Z < 3.0$ two samples are significantly different,

$3.0 < Z$ two samples are highly significantly different.

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