Supplement of Atmos. Chem. Phys., 22, 10721–10749, 2022
https://doi.org/10.5194/acp-22-10721-2022-supplement

Atmospheric Chemistry and Physics
Open Access
EGU

Supplement of

Analysis of regional CO_2 contributions at the high Alpine observatory Jungfraujoch by means of atmospheric transport simulations and $\delta^{13}C$

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Table S1. File description and categorisation of EDGARv4.3-fuel inventory for FLEXPART-COSMO.

File	Fuel [a]	IPCC [b]	IPCC [b]	Time Factor [c]	Annual Factor [d]
data_CO2 (0.1x0.1.nc, grmnht)					
fossil fuel, coal brown ("bituminous")					
v43_2010_CO2_ENE_coal_brown	coal_brown	1.A.1.a	ENE	A	coal
v43_2010_CO2_INS_coal_brown	coal_brown	1.A.2+6.C.D	IND	B	coal
v43_2010_CO2_RTNE_Coal_brown	coal_brown	1.A.1.b.c.r	REF_TRF	D	coal
v42_FT2010_CO2_RCO_COAL_brown	coal_brown	1.A.4	RCO	C	coal
fossil fuel, coal hard ("anthracite")					
v43_2010_CO2_ENE_coal_hard	coal_hard	1.A.1.a	ENE	A	coal
v43_2010_CO2_INS_coal_hard	coal_hard	1.A.2+6.C.D	IND	B	coal
v43_2010_CO2_RTNE_coal_hard_ff	coal_hard	1.A.1.b.c.r	REF_TRF	D	coal
v42_FT2010_CO2_RCO_coal_hard	coal_hard	1.A.4	RCO	C	coal
fossil fuel, peat					
v43_2010_CO2_ENE_coal_peat	coal_peat	1.A.1.a	ENE	A	coal
v43_2010_CO2_INS_coal_peat	coal_peat	1.A.2+6.C.D	IND	B	coal
v43_2010_CO2_RTNE_coal_peat	coal_peat	1.A.1.b.c.r	REF_TRF	D	coal
v42_FT2010_CO2_RCO_coal_peat	coal_peat	1.A.4	RCO	C	coal
fossil fuel, gas derived					
v43_2010_CO2_ENE_gas_der	gas_derived	1.A.1.a	ENE	A	gas
v43_2010_CO2_INS_gas_der	gas_derived	1.A.2+6.C.D	IND	B	gas
v43_2010_CO2_RTNE_gas_der_c	gas_derived	1.A.1.b.c.r	REF_TRF	D	gas
v42_FT2010_CO2_RCO_gas_derived	gas_derived	1.A.4	RCO	C	gas
fossil fuel, gas natural					
v43_2010_CO2_ENE_gas_nat	gas_natural	1.A.1.a	ENE	A	gas
v43_2010_CO2_INS_gas_nat	gas_natural	1.A.2+6.C.D	IND	B	gas
v43_2010_CO2_RTNE_gas_nat	gas_natural	1.A.1.b.c.r	REF_TRF	D	gas
v42_FT2010_CO2_RCO_gas_natural	gas_natural	1.A.4	RCO	C	gas
fossil fuel, oil heavy					
v43_2010_CO2_ENE_oil_heavy	oil_heavy	1.A.1.a	ENE	A	oil
v43_2010_CO2_INS_oil_heavy	oil_heavy	1.A.2+6.C.D	IND	B	oil
v43_2010_CO2_PRO_oil_heavy	oil_heavy	1.B.2.a.b.c	PRO	D	oil
v43_2010_CO2_RTNE_oil_heavy	oil_heavy	1.A.1.b.c.r	REF_TRF	D	oil
v43_2010_CO2_TRO_oil_heavy	oil_heavy	1.A.3.b	TRO	F	oil
v43_EM_CO2_2010_TNG_CO2_2010_TNG	oil_heavy	1.A.3.c.e	TNR_Other	I	oil
v42_FT2010_CO2_excl_short-cycle_org_C_2010_IPCC_1A3d	oil_heavy	1.A.3.d.+1.C.2	TNR_Ship	G	oil
v42_FT2010_CO2_RCO_oil_heavy	oil_heavy	1.A.4	RCO	C	oil
fossil fuel, oil light					
v43_2010_CO2_ENE_oil_light	oil_light	1.A.1.a	ENE	A	oil
v43_2010_CO2_INS_oil_light	oil_light	1.A.2+6.C.D	IND	B	oil
v43_2010_CO2_PRO_oil_light	oil_light	1.B.2.a.b.c	PRO	D	oil
v43_2010_CO2_RTNE_oil_light	oil_light	1.A.1.b.c.r	REF_TRF	D	oil
v43_2010_CO2_TRO_oil_light	oil_light	1.A.3.b	TRO	F	oil
v42_FT2010_CO2_excl_short-cycle_org_C_2010_IPCC_1A3a	oil_light	1.A.3.a.+1.C.1	TNR_Aviation	H	oil
v42_FT2010_CO2_excl_short-cycle_org_C_2010_IPCC_7A	oil_light	7.A	FFF	n.a.	oil
v42_FT2010_CO2_RCO_OIL_light	oil_light	1.A.4	RCO	C	oil
fossil fuel, oil mixed					
v43_2010_CO2_PRO_oil_lght+hvy+gas_VAF_transmit	oil_mixed	1.B.2.a.b.c	PRO	D	oil
solid waste, non-biogenic*					
v43_2010_CO2_ENE_solid_non_bio_waste	solid_waste_non_bio	1.A.1.a	ENE	A	n.a.
v43_2010_CO2_INS_solid_non_bio_f_solid_wst_nonbio	solid_waste_non_bio	1.A.2+6.C.D	IND	B	n.a.
v43_2010_CO2_RTNE_solid_non_bio_f	solid_waste_non_bio	1.A.1.b.c.r	REF_TRF	D	n.a.
v43_2010_CO2_RCO_solid_non_bio_f_solidwaste	solid_waste_non_bio	1.A.4	RCO	C	n.a.
non-fuel emissions*					
v42_FT2010_CO2_excl_short-cycle_org_C_2010_IPCC_2A	cement	2.A	NMM	B	n.a.
v42_FT2010_CO2_excl_short-cycle_org_C_2010_IPCC_2B_3	other	2.B.E.F.G+3	CHE	B	n.a.
v42_FT2010_CO2_excl_short-cycle_org_C_2010_IPCC_2C	other	2.C	IRO	B	n.a.
v42_FT2010_CO2_excl_short-cycle_org_C_2010_IPCC_4C_4D	agri	4.C._4.D	AGS	L	n.a.
wildfires*					

v42_FT2010_CO2_BMB_FR_solid_biomass	biomass_wildfires	n.a.	BMB	n.a.	n.a.
v42_FT2010_CO2_BMB_peat	peat_wildfires	n.a.	BMB	n.a.	n.a.
data_CO2x (0.1x0.1.nc, grmnht)					
biogas					
v43_2010_CO2_ENE_gas_bio	biogas (bio, gas)	1.A.1.a	ENE	A	bio
v43_2010_CO2_INS_gas_bio	biogas (bio, gas)	1.A.2+6.C.D	IND	B	bio
v43_2010_CO2_RCO_bio_gas	biogas (bio, gas)	1.A.4	RCO	C	bio
v43_2010_CO2_RTNE_gas_bio	biogas (bio, gas)	1.A.1.b.c.r	REF_TRF	D	bio
biofuels liquid					
v43_2010_CO2_ENE_liquid_bio	bioliquid (bio, liquid)	1.A.1.a	ENE	A	bio
v43_2010_CO2_INS_liquid_bio	bioliquid (bio, liquid)	1.A.2+6.C.D	IND	B	bio
v43_2010_CO2_RCO_bio_liquid	bioliquid (bio, liquid)	1.A.4	RCO	C	bio
biomass					
v43_2010_CO2_ENE_solid_bio	biomass (bio, solid)	1.A.1.a	ENE	A	n.a.
v43_2010_CO2_INS_solid_bio_f	biomass (bio, solid)	1.A.2+6.C.D	IND	B	n.a.
v43_2010_CO2_RCO_bio_solid	biomass (bio, solid)	1.A.4	RCO	C	n.a.
v43_2010_CO2_RTNE_solid_bio_f	biomass (bio, solid)	1.A.1.b.c.r	REF_TRF	D	n.a.
v42_FT2010_CO2_short_cycle_org_C_AWB	biomass (bio, solid)	4.F	AWB	L	n.a.

[a)] compare Janssens-Maenhout et al., (2019); Karstens (2019); Vardag et al. (2015 and 2016) *solid waste, agri and wildfires are excluded from our FLEXPART-COSMO analysis, in order to use the same categorization as for STILT-ECMWF by the ICOS Carbon Portal

[b)] compare Janssens-Maenhout et al. (2019)

[c)] GNFR categories for time factors as summarized in Table S3; compare Kuenen et al. (2014).

[d)] Categories based on BP Statistics (bp, 2019) as summarized in Table S2; n.a. = based on 2010 and no further factor applied.

Table S2. Annual scaling factors based on BP (2019).

Factor		2009	2010	2011	2012	2013	2014	2015	2016	2017
oil										
	Switzerland	1.078	1	0.970	0.988	1.035	0.928	0.942	0.893	0.918
	Austria	0.961	1	0.930	0.927	0.940	0.915	0.919	0.937	0.954
	France	1.035	1	0.979	0.951	0.939	0.912	0.908	0.902	0.906
	Germany	0.993	1	0.969	0.965	0.983	0.958	0.955	0.975	0.996
	Italy	1.026	1	0.964	0.901	0.822	0.777	0.812	0.818	0.824
gas										
	Switzerland	0.895	1	0.887	0.972	1.024	0.887	0.948	0.996	1.000
	Austria	0.919	1	0.941	0.897	0.853	0.781	0.829	0.868	0.944
	France	0.902	1	0.868	0.897	0.911	0.765	0.822	0.898	0.903
	Germany	0.959	1	0.918	0.921	0.965	0.839	0.874	0.964	1.019
	Italy	0.939	1	0.938	0.902	0.843	0.745	0.813	0.853	0.904
coal										
	Switzerland	0.997	1	0.924	0.833	0.897	0.918	0.839	0.771	0.742
	Austria	0.850	1	1.027	0.954	0.970	0.889	0.961	0.891	0.923
	France	0.934	1	0.849	0.961	1.005	0.750	0.728	0.709	0.805
	Germany	0.931	1	1.016	1.045	1.075	1.033	1.021	0.993	0.928
	Italy	0.904	1	1.121	1.149	0.990	0.965	0.902	0.803	0.704
bio										
	Switzerland	na	na	na	na	na	na	na	na	na
	Austria	0.954	1	0.997	0.997	0.958	0.843	0.975	1.073	1.000
	France	1.026	1	0.910	1.051	1.073	1.135	1.128	1.063	1.153
	Germany	0.807	1	0.983	0.921	0.981	1.104	1.018	1.030	1.051
	Italy	0.960	1	0.754	0.400	0.587	0.659	0.667	0.674	0.808

Factors are applied country specific when data are available for the entire domain, although only factors for Switzerland and source regions bordering with Switzerland are provided in this table. "coal", "gas" and "oil" refer to data on coal, gas and oil consumption per country and year. The liquid biofuel consumption is included in oil consumption data; "bio" refers to the bioliquid production per country and year under the assumption that production equals consumption. The bioliquid data are also used for biogas, as no other information is available.; n.a. = based on 2010 and no further factor applied.

Table S3. Time factors to account for seasonal, weekly and diurnal cycles.

Factor (GNFR)	seasonal (Jan-Dec)	weekly (Mo-Sun)	diurnal (1-24h)
A, Public Power	1.2/1.15/1.05/1/0.9/0.85/0.8/0.875/0.95/1/1.075/1.15	1.06/1.06/1.06/1.06/1.06/0.85/0.85	0.79/0.72/0.72/0.71/0.74/0.8/0.92/1.08/1.19/1.22/1.21/1.21/ 1.17/1.15/1.14/1.13/1.1/1.07/1.04/1.02/1.02/1.01/0.96/0.88
B, Industry	1.1/1.075/1.05/1/0.95/0.9/0.93/0.95/0.97/1/1.025/1.05	1.08/1.08/1.08/1.08/1.08/0.8/0.8	0.75/0.75/0.78/0.82/0.88/0.95/1.02/1.09/1.16/1.22/1.28/1.3/ 1.22/1.24/1.25/1.16/1.08/1.01/0.95/0.9/0.85/0.81/0.78/0.75
C, Other Stationary Combustion	1.7/1.5/1.3/1/0.7/0.4/0.2/0.4/0.7/1.05/1.4/1.65	1.08/1.08/1.08/1.08/1.08/0.8/0.8	0.38/0.36/0.36/0.36/0.37/0.5/1.19/1.53/1.57/1.56/1.35/1.16/ 1.07/1.06/1/0.98/0.99/1.12/1.41/1.52/1.39/1.35/1/0.42
D, Fugitives	1.2/1.2/1.2/0.8/0.8/0.8/0.8/0.8/0.8/1.2/1.2/1.2	1	1
F, Road Transport	0.88/0.92/0.98/1.03/1.05/1.06/1.01/1.02/1.06/1.05/1.01/0.93	1.02/1.06/1.08/1.1/1.14/0.81/0.79	0.19/0.09/0.06/0.05/0.09/0.22/0.86/1.84/1.86/1.41/1.24/1.2/ 1.32/1.44/1.45/1.59/2.03/2.08/1.51/1.06/0.74/0.62/0.61/0.44
G, Shipping	1	1	1
H, Aviation	1	1	1
I, OffRoad Transport	1	1	1
L, Agri Other	0/2/4.75/2.9/0.5/0.4/0.2/0.5/0.75/0/0/0	1	1

TNO-MACC GNFR Time Factors as of September 2018, e.g. as described in Kuenen et al. (2014).

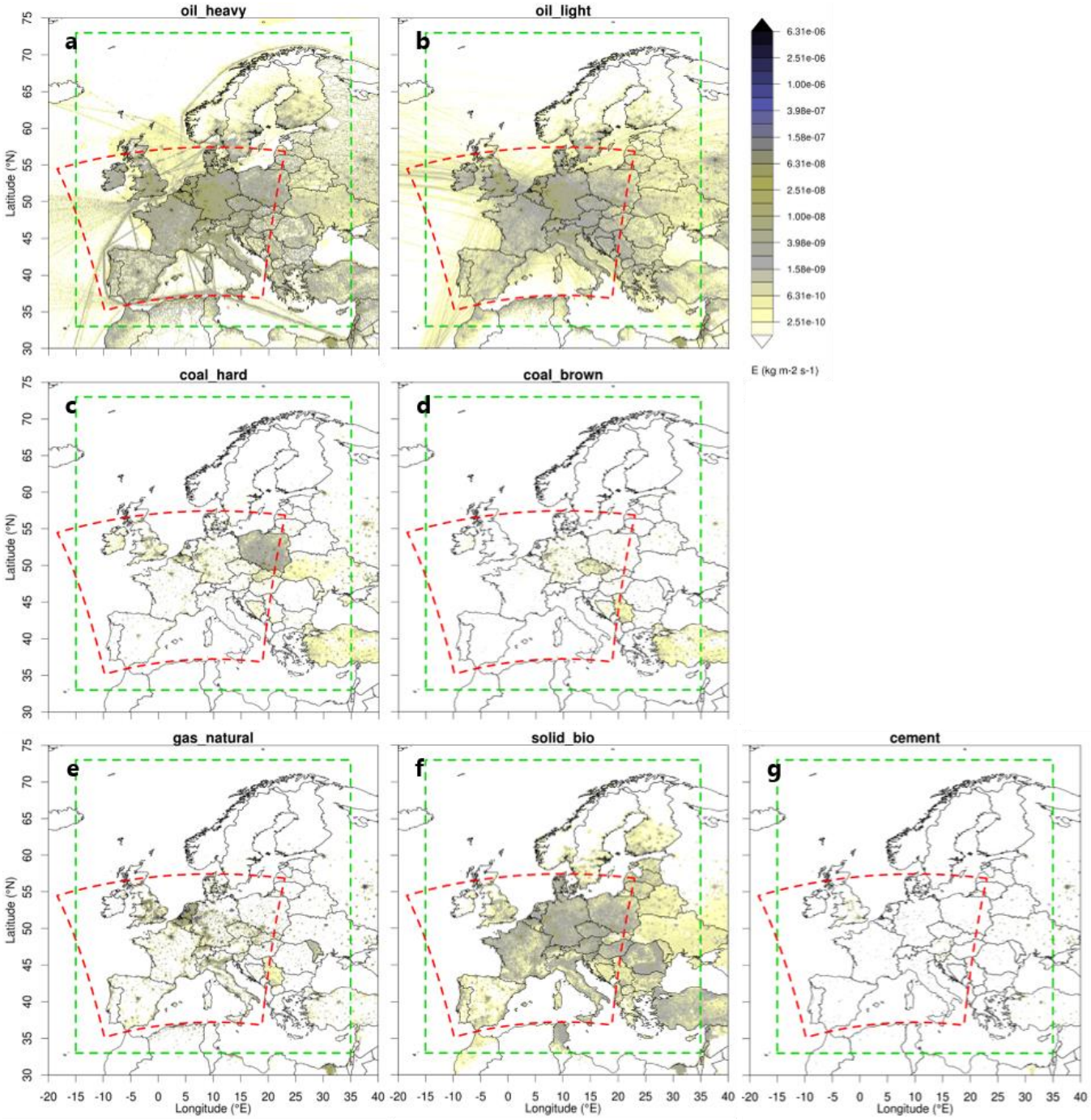


Figure S1. EDGARv4.3 anthropogenic emissions maps for the base year 2010 for seven prominent categories concerning CO_2 contributions at JFJ: **a)** oil_heavy, **b)** oil_light, **c)** coal_hard, **d)** coal_brown, **e)** gas_natural, **f)** solid biomass, and **g)** cement production, with indication of FLEXPART-COSMO (red) and STILT-ECMWF (green) domain boundaries as used in our study.

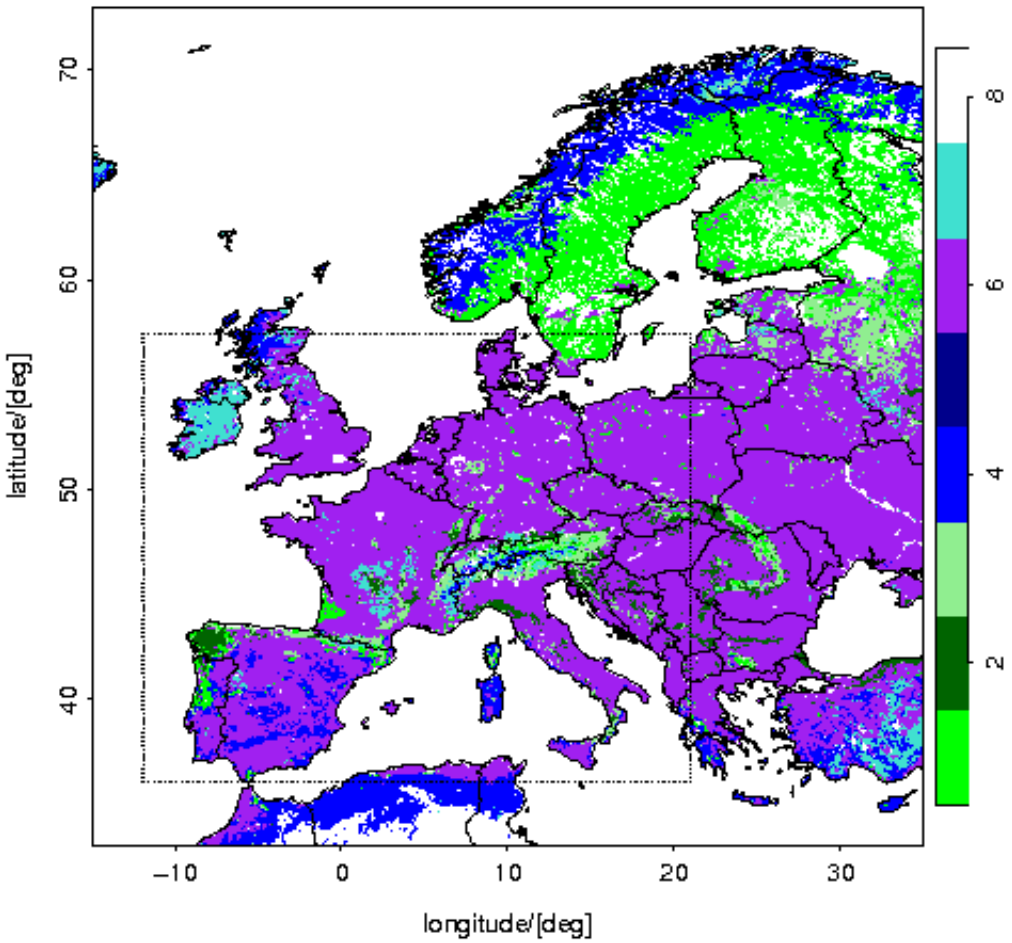


Figure S2. Dominant vegetation type (VT) per grid. These VTs include 1) evergreen, 2) deciduous, and 3) mixed forest, 4) shrubland (including open and closed shrubland), 5) savanna (savannas and woody savannas), 6) cropland, 7) grass- land (grassland, cropland/natural vegetation mosaic, and barren or sparsely vegetated), and 8) others. VT 5 (savanna) is not present within the domain as it is not a European ecosystem.

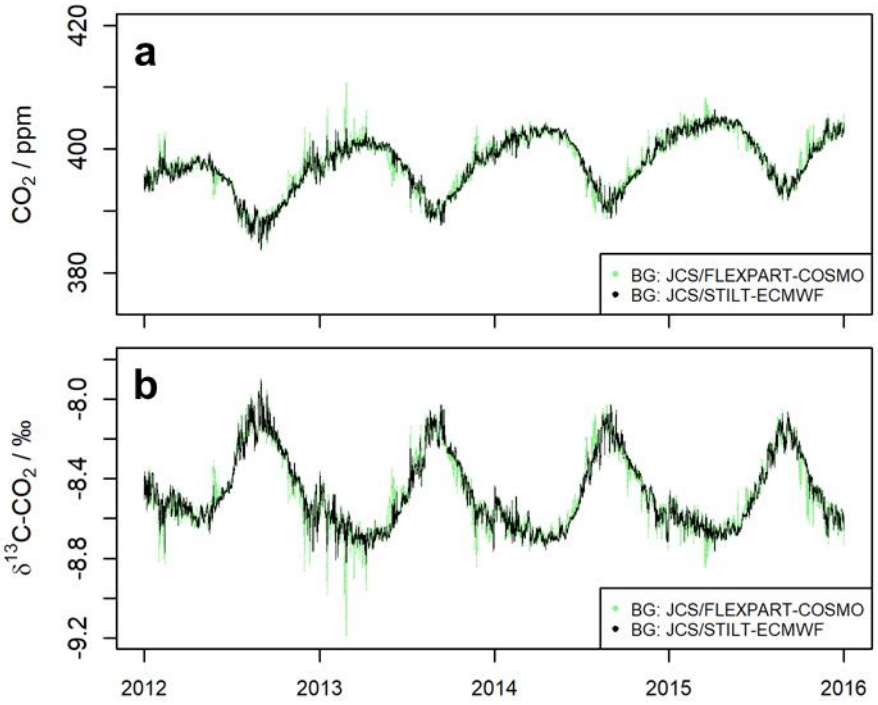


Figure S3. Time series of **a)** Jena CarboScope (JCS) CO_2 concentration background (f_b), **b)** $\delta^{13}C$-CO_2 background based on f_b and yearly fits (based on strategy used by Vardag et al. 2016)

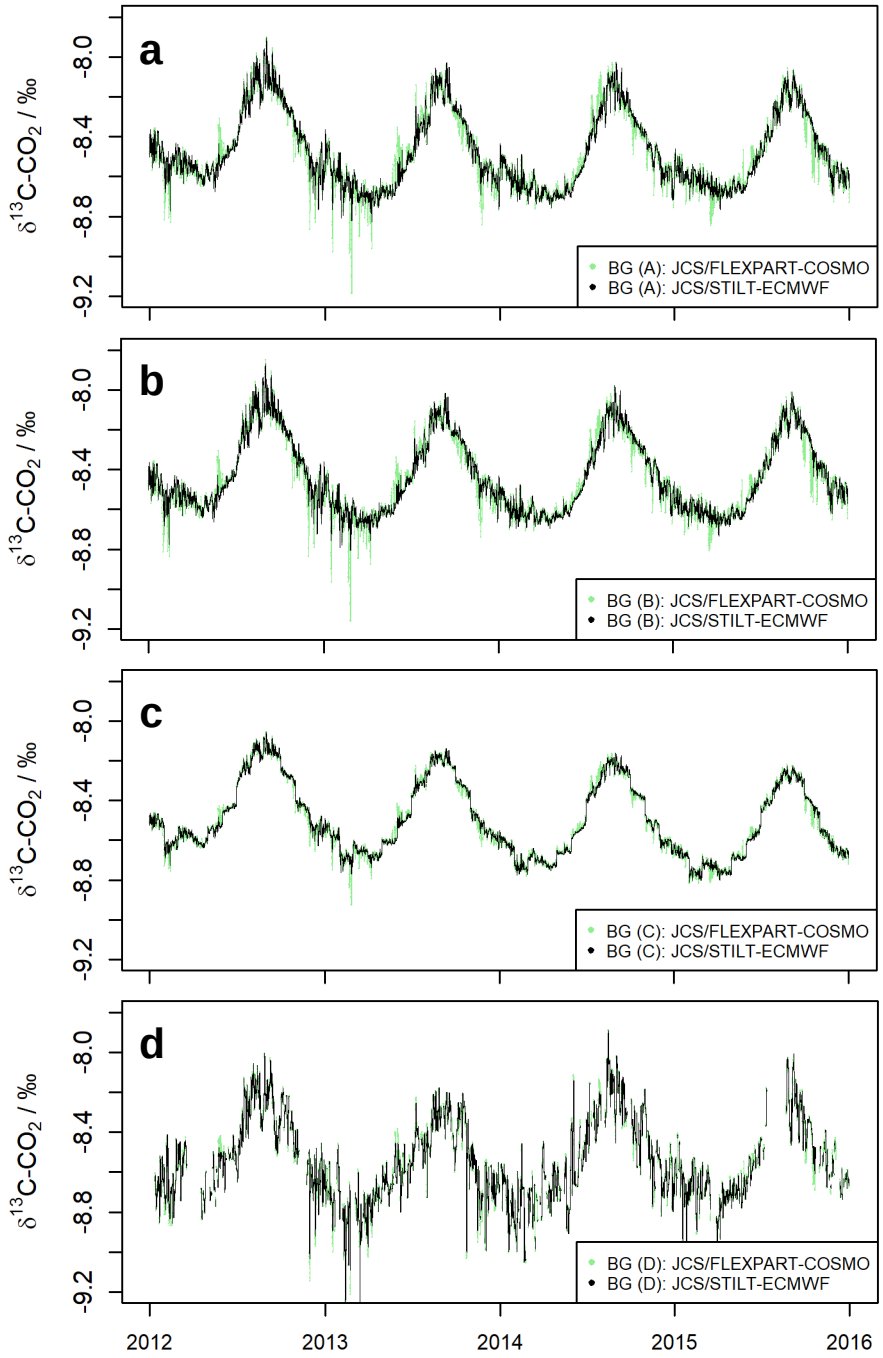


Figure S4. Time series of $\delta^{13}C\text{-}CO_2$ background **a)** based on f_b and yearly fits (strategy used by Vardag et al. 2016, and equivalent to Fig. S3b), b) based on f*b* and moving fits (12 months window), c) based on f*b* and multi-annual monthly ratios of $\delta^{13}C\text{-}CO_2$ to CO_2, **d)** based on f*b* and daily ratios of $\delta^{13}C\text{-}CO_2$ to CO_2 at 5-6 AM local time. **a)-c)** are based on flask data provided by MPI-BGC, **d)** is based on QCLAS data.

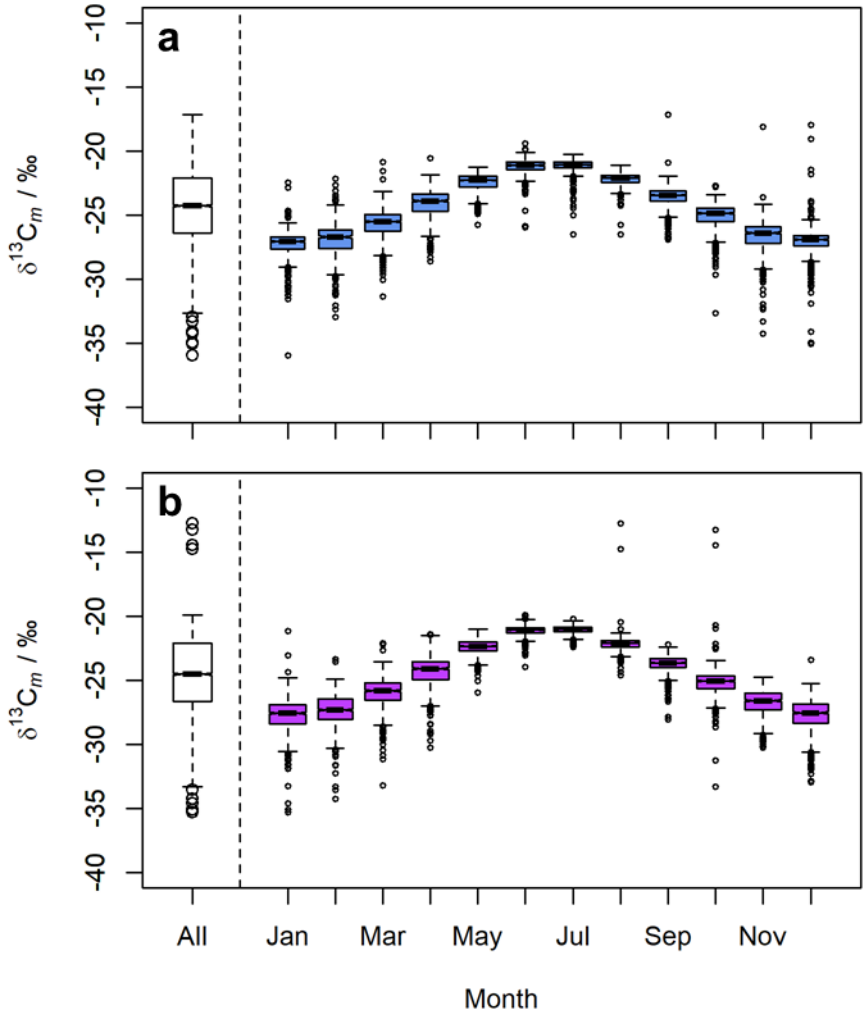


Figure S5 Multi-annual boxplots of model-based mixed source signatures (**a**) ($\delta^{13}C_m$) for 2012–2015 (3-hourly data). **a)** FLEXPART-COSMO, **b)** STILT-ECMWF. The outliers indicate events of particular source compositions.

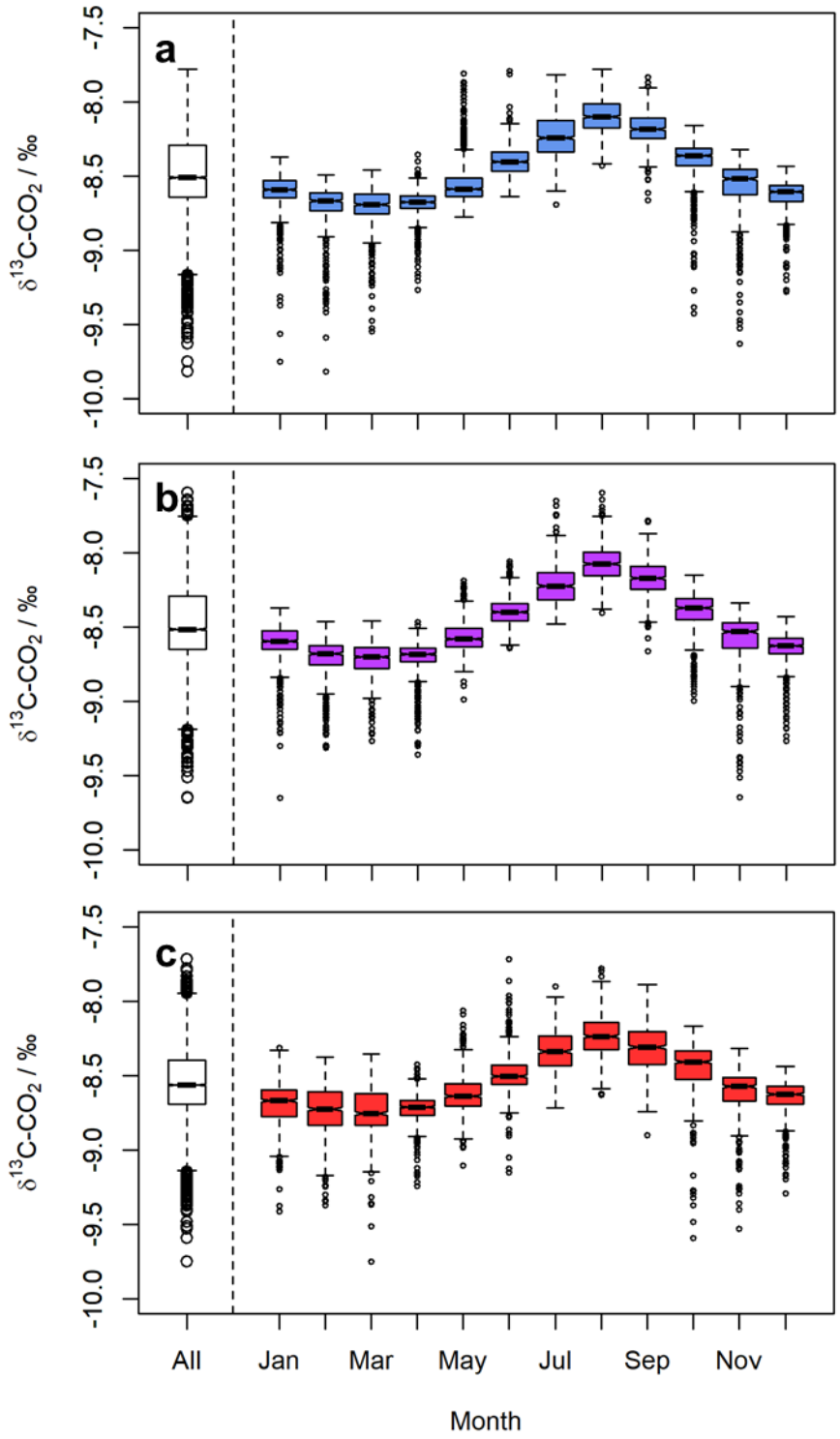


Figure S6. Multi-annual boxplots of atmospheric $\delta^{13}C\text{-}CO_2$ for 2012–2015 (3-hourly data). **a)** model-based, FLEXPART-COSMO, **b)** model-based**,** STILT-ECMWF, **c)** observations (QCLAS). The outliers indicate intense regional events.

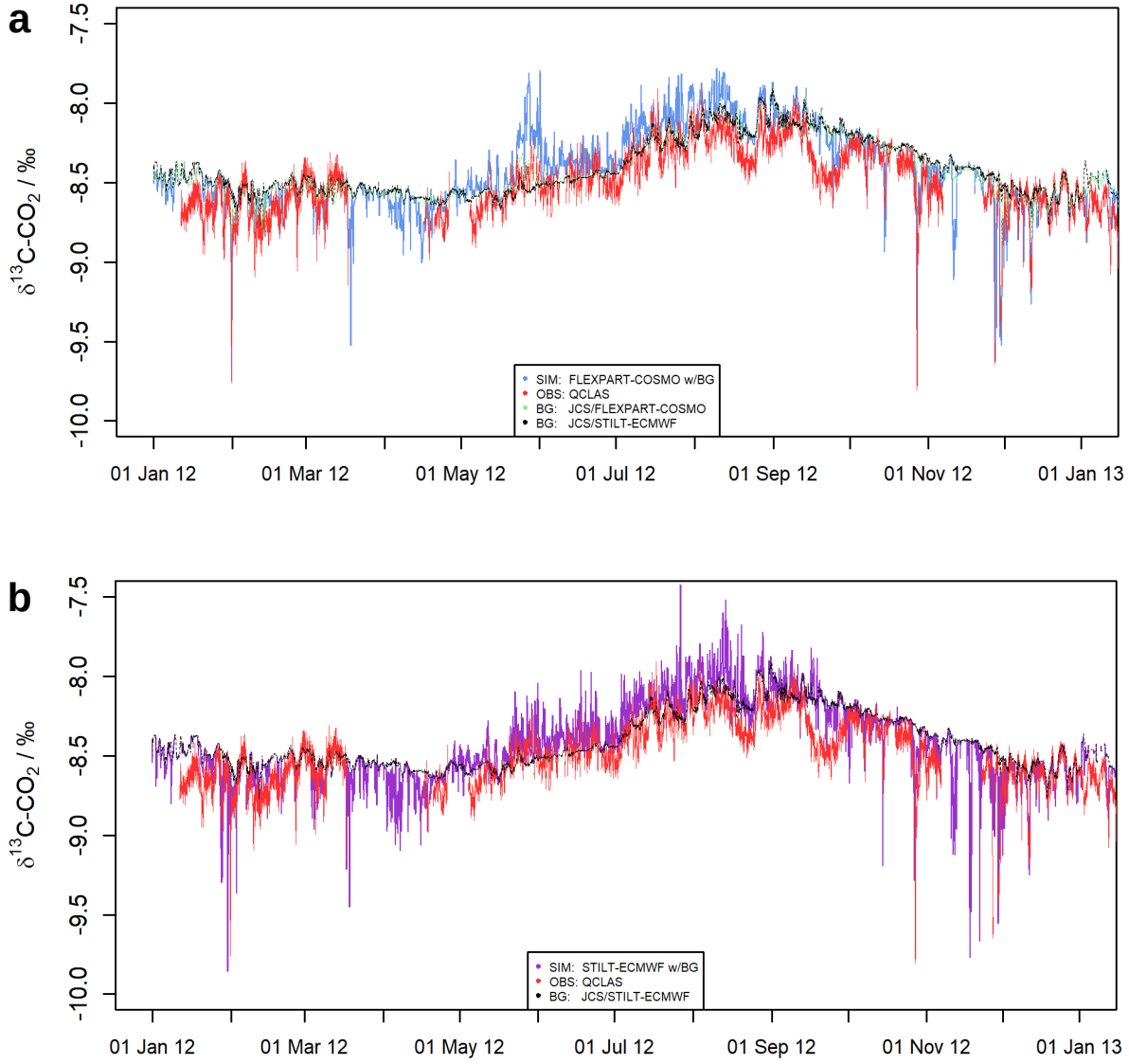


Figure S7. Zoom into 2012 (see caption Figure 6, main text).

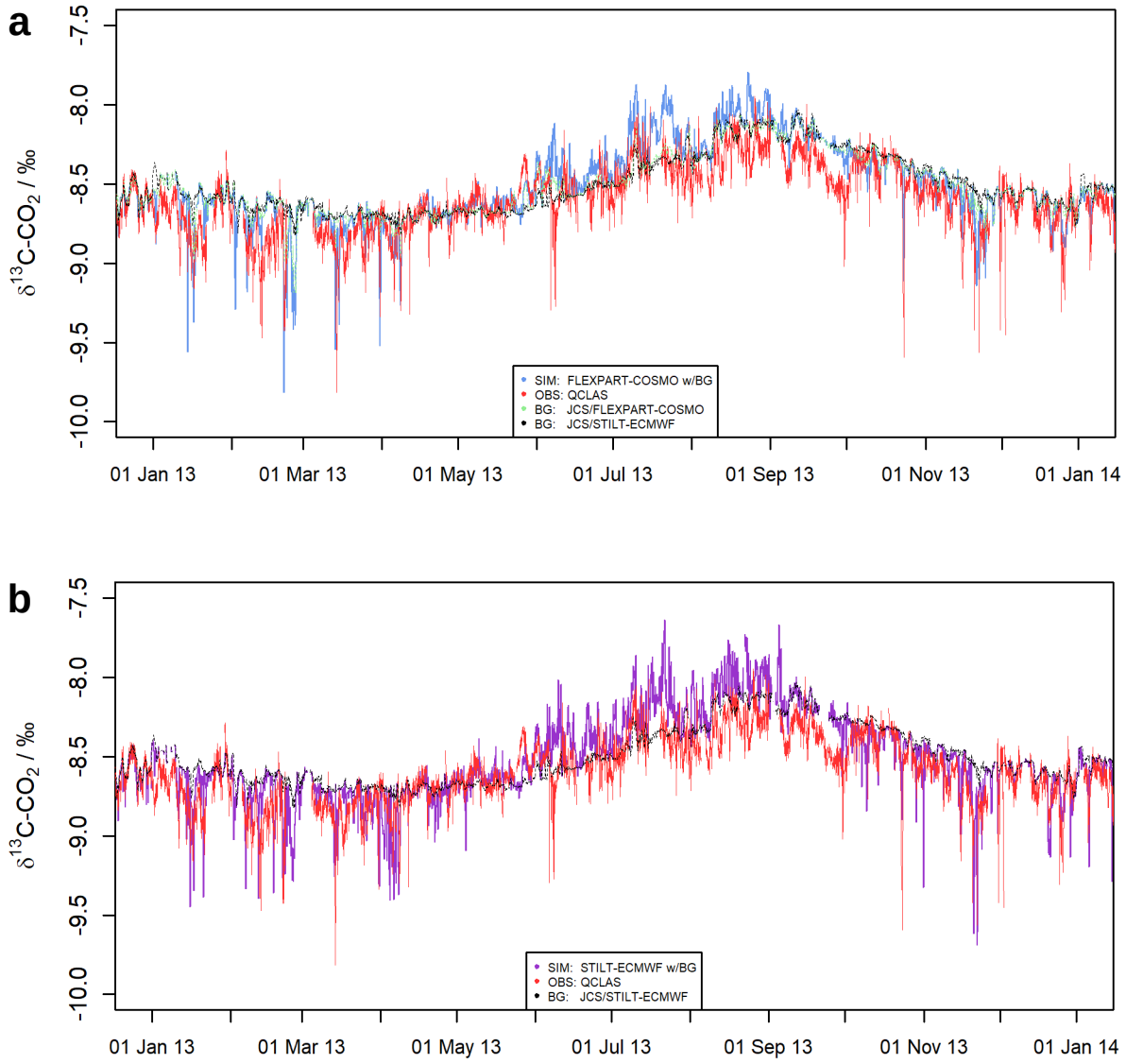


Figure S8. Zoom into 2013 (see caption Figure 6, main text).

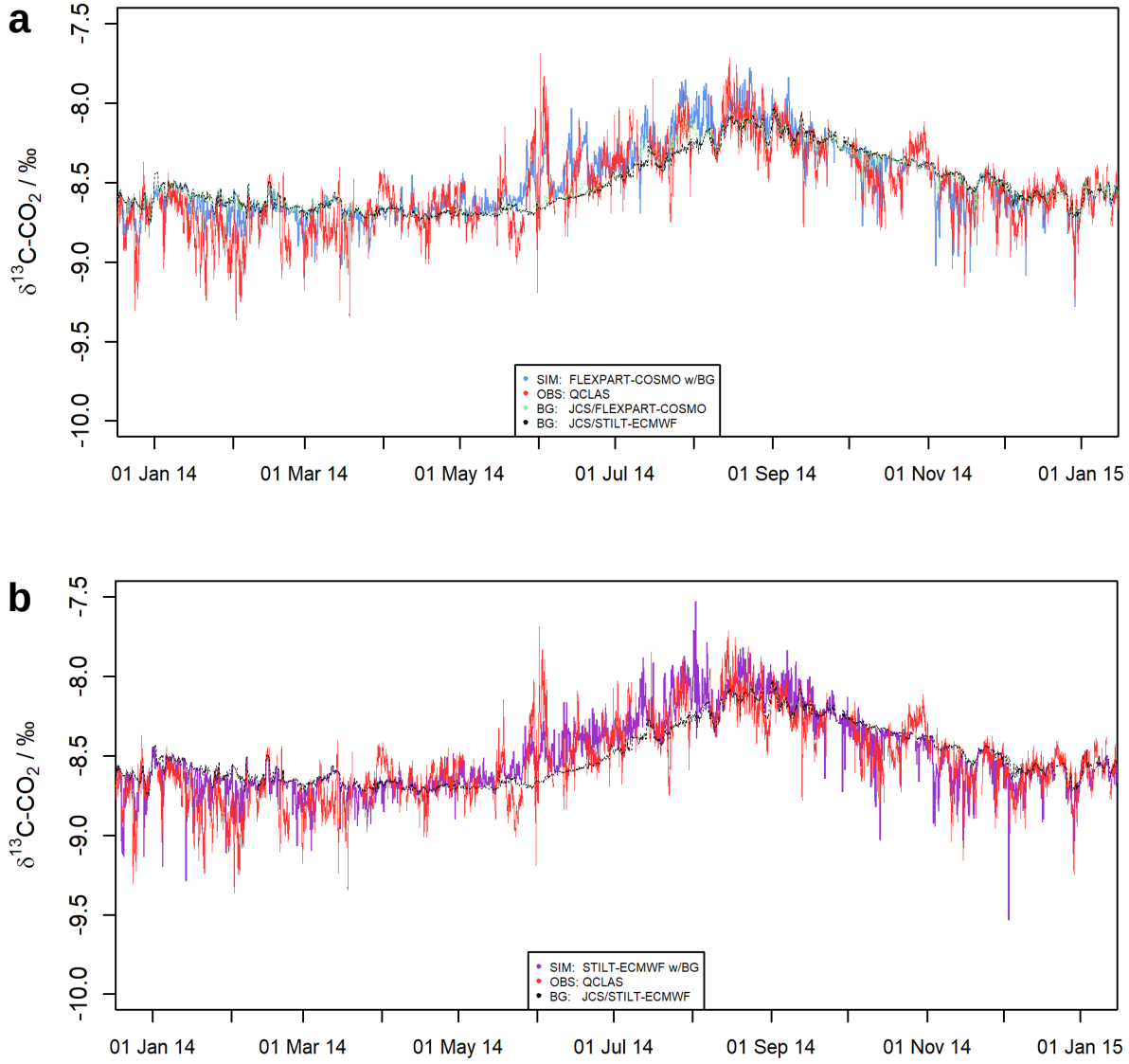


Figure S9. Zoom into 2014 (see caption Figure 6, main text).

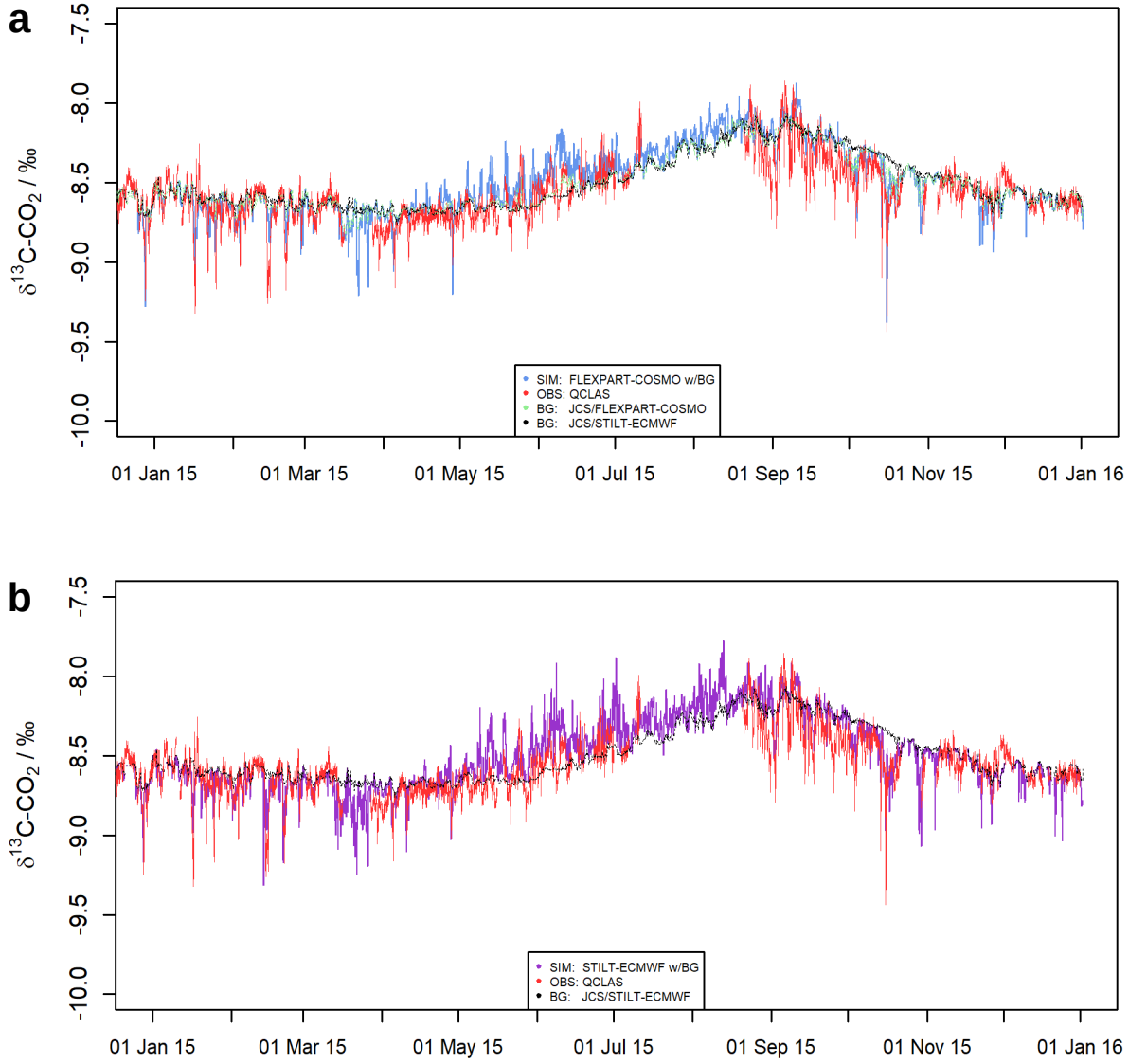
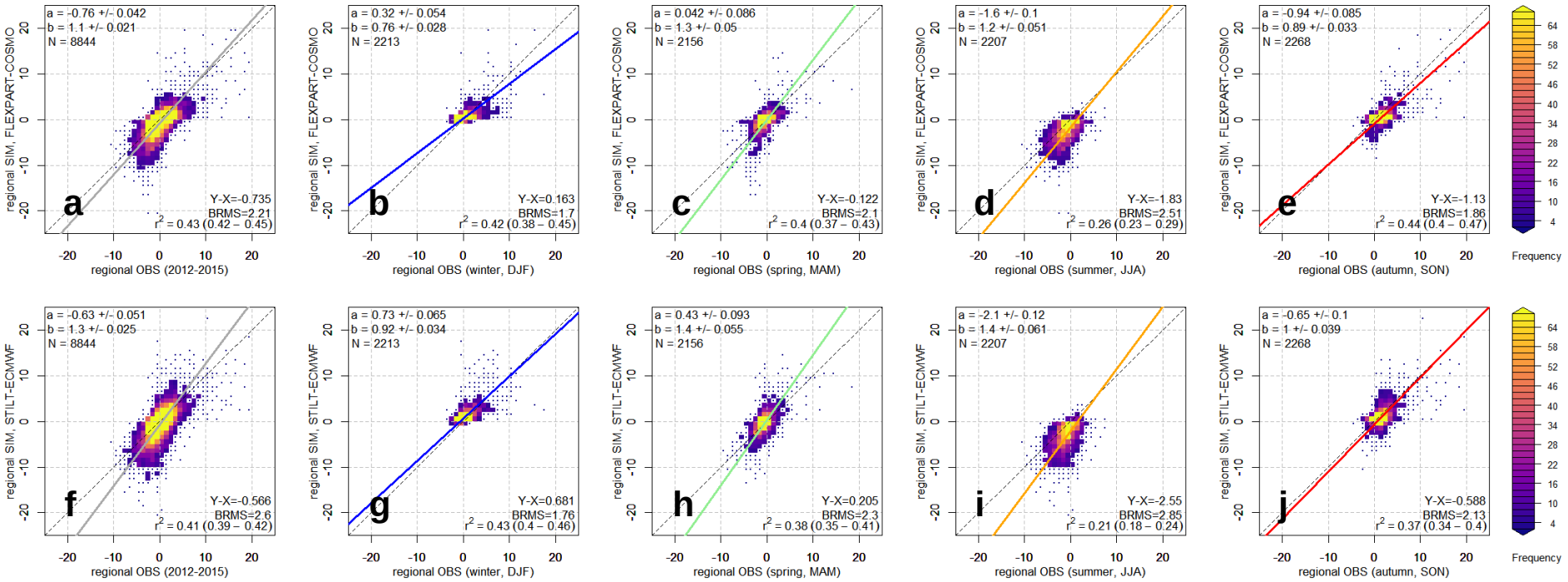


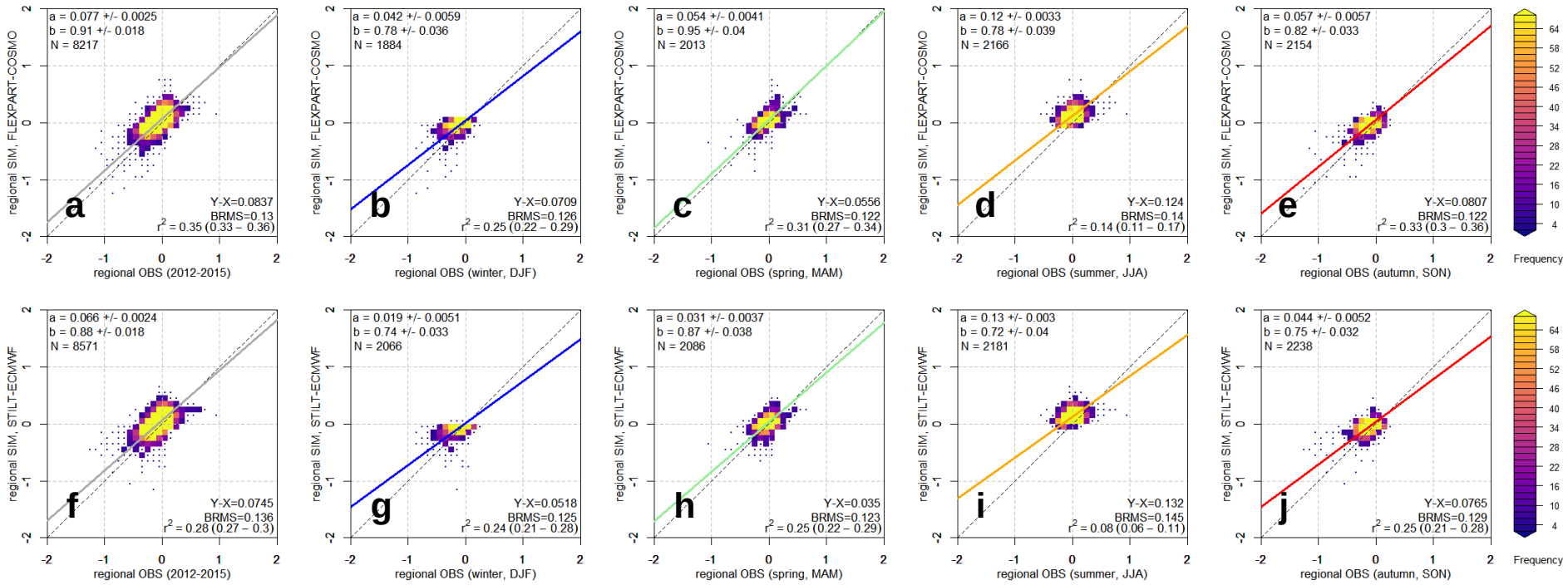
Figure S10. Zoom into 2015 (see caption Figure 6, main text).

Table S4. Results from the CO_2 sensitivity analysis for JJA (2012-2015) and the effect on the regional $\delta^{13}C$-CO_2 simulation.

		FLEXPART-COSMO (2012-2015, JJA)						STILT-ECMWF (2012-2015, JJA)				
CO_2 regional	**base**	**sc1 (anthr)**	**sc2 (nee)**	**sc3 (gee)**	**sc4 (resp)**	**sc5 (trans)**	**base**	**sc1 (anthr)**	**sc2a (nee)**	**sc3 (gee)**	**sc4 (resp)**	**sc5 (trans)**
scaling factor, slope		*3.14*	*0.80*	*0.79*	*2.08*	*0.82*		*3.73*	*0.72*	*0.74*	*1.98*	*0.74*
scaling factor, intercept		*0.02*	*1.04*	*0.45*	*-0.88*	*-0.88*		*-0.11*	*1.22*	*0.49*	*-0.56*	*1.54*
r^2 (low-high)	0.26 (0.23-0.29)	0.14 (0.11-0.17)	0.26 (0.23-0.29)	0.28 (0.25-0.31)	0.24 (0.21-0.28)	0.26 (0.26-0.29)	0.21 (0.18-0.24)	0.12 (0.10-0.15)	0.21 (0.18-0.24)	0.23 (0.20-0.26)	0.21 (0.18-0.24)	0.21 (0.18-0.24)
bias, Y-X (ppm)	-1.83	-0.02	-1.05	-0.48	0.87	-1.28	-2.55	0.11	-1.21	-0.48	0.56	-1.56
BRMS (ppm)	2.51	2.66	2.16	2.03	2.19	2.23	2.85	3.07	2.28	2.16	2.56	2.35
slope± err	1.20± 0.05	1.10± 0.05	0.93± 0.04	0.83± 0.03	0.94± 0.04	1.00± 0.04	1.40± 0.06	1.30± 0.07	0.94± 0.04	0.88± 0.04	1.20± 0.05	1.00± 0.05
$\delta^{13}C$ regional	**Base**	**sc1 (anthr)**	**sc2a (nee)**	**sc3 (gee)**	**sc4 (resp)**	**sc5 (trans)**	**base**	**sc1 (anthr)**	**sc2a (nee)**	**sc3 (gee)**	**sc4 (resp)**	**sc5 (trans)**
r^2 (low-high)	0.14 (0.11-0.17)	0.09 (0.07-0.12)	0.14 (0.12-0.17)	0.16 (0.13-0.19)	0.19 (0.16-0.22)	0.14 (0.12-0.17)	0.08 (0.06-0.11)	0.08 (0.06-0.10)	0.09 (0.07-0.12)	0.12 (0.10-0.15)	0.15 (0.12-0.17)	0.08 (0.06-0.11)
bias, Y-X (‰)	0.12	0.06	0.10	0.08	0.03	0.11	0.13	0.04	0.09	0.06	0.03	0.10
BRMS (‰)	0.14	0.14	0.13	0.13	0.13	0.13	0.14	0.15	0.13	0.13	0.13	0.14
slope± err	0.78± 0.04	0.73± 0.04	0.64± 0.03	0.59± 0.03	0.64± 0.03	0.67± 0.03	0.72± 0.04	0.80± 0.05	0.50± 0.03	0.46± 0.02	0.65± 0.03	0.53± 0.03



Copy of Figure 5 (main text). Heatmaps for CO_2.*regional* simulations (SIM) using FLEXPART-COSMO (**a-e**) and STILT-ECMWF (**f-j**), in comparison to regional components of observations (OBS) for 2012–2015, full year and per seasons, on 3-hourly time resolution. The STILT-ECMWF-based JCS background is subtracted from the observations to derive the regional component. The weighted least squares regression takes into account uncertainties in both data sets.



Copy of Figure 10 (main text). Heatmaps of model-based regional $\delta^{13}C\text{-}CO_2$ (SIM) vs. observation (OBS) (3-hourly data), for FLEXPART-COSMO (**a-e**) and STILT-ECMWF (**f-j**), during 2012-2015, for the full year (grey), and per season (DJF (blue), MAM (green), JJA (orange), SON (red)). Uncertainties in x- and y-axes are taken into account in the weighted least squares regression applied here.

References

bp: Statistical Review of World Energy (Version 2019), [online] Available from: https://www.bp.com/en/global/corporate/energy-economics/statistical-review-of-world-energy.html (Accessed 26 June 2019), 2019.

Janssens-Maenhout, G., Crippa, M., Guizzardi, D., Muntean, M., Schaaf, E., Dentener, F., Bergamaschi, P., Pagliari, V., Olivier, J. G. J., Peters, J. A. H. W., Van Aardenne, J. A., Monni, S., Doering, U., Roxana Petrescu, A. M., Solazzo, E. and Oreggioni, G. D.: EDGAR v4.3.2 Global Atlas of the three major greenhouse gas emissions for the period 1970-2012, Earth Syst. Sci. Data, 11(3), 959–1002, doi:10.5194/essd-11-959-2019, 2019.

Karstens, U.: Global anthropogenic CO_2 emissions for 2006-2019 based on EDGARv4.3 and BP statistics 2019 (Version 2.0), ICOS ERIC - Carbon Portal, [online] Available from: https://doi.org/10.18160/Y9QV-S113 (Accessed 26 June 2019), 2019.

Kuenen, J. J. P., Visschedijk, A. J. H., Jozwicka, M. and Denier Van Der Gon, H. A. C.: TNO-MACC-II emission inventory; A multi-year (2003-2009) consistent high-resolution European emission inventory for air quality modelling, Atmos. Chem. Phys., 14(20), 10963–10976, doi:10.5194/acp-14-10963-2014, 2014.

Vardag, S. N., Gerbig, C., Janssens-Maenhout, G. and Levin, I.: Estimation of continuous anthropogenic CO_2: Model-based evaluation of CO_2, CO, $\delta^{13}C(CO_2)$ and $\Delta^{14}C(CO_2)$ tracer methods, Atmos. Chem. Phys., 15(22), 12705–12729, doi:10.5194/acp-15-12705-2015, 2015.

Vardag, S. N., Hammer, S. and Levin, I.: Evaluation of 4 years of continuous $\delta^{13}C(CO_2)$ data using a moving Keeling plot method, Biogeosciences, 13(14), 4237–4251, doi:10.5194/bg-13-4237-2016, 2016.