

***Supplement of***

**Analysis of regional CO<sub>2</sub> contributions at the high Alpine observatory Jungfraujoch by means of atmospheric transport simulations and δ<sup>13</sup>C**

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

File	Fuel <sup>a)</sup>	IPCC <sup>b)</sup>	IPCC <sup>b)</sup>	Time Factor <sup>c)</sup>	Annual Factor <sup>d)</sup>
<b>data_CO2 (0.1x0.1.nc, grmnht)</b>					
<b>fossil fuel, coal brown ("bituminous")</b>					
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
<b>fossil fuel, coal hard ("anthracite")</b>					
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
<b>fossil fuel, peat</b>					
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
<b>fossil fuel, gas derived</b>					
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
<b>fossil fuel, gas natural</b>					
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
<b>fossil fuel, oil heavy</b>					
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
<b>fossil fuel, oil light</b>					
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
<b>fossil fuel, oil mixed</b>					
v43_2010_CO2_PRO_oil_lght+hvy+gas_VAF_transmit	oil_mixed	1.B.2.a.b.c	PRO	D	oil
<b>solid waste, non-biogenic*</b>					
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.
<b>non-fuel emissions*</b>					
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.
<b>wildfires*</b>					

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.
<b>data_CO2x (0.1x0.1.nc, grmnht)</b>					
<b>biogas</b>					
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
<b>biofuels liquid</b>					
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
<b>biomass</b>					
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).

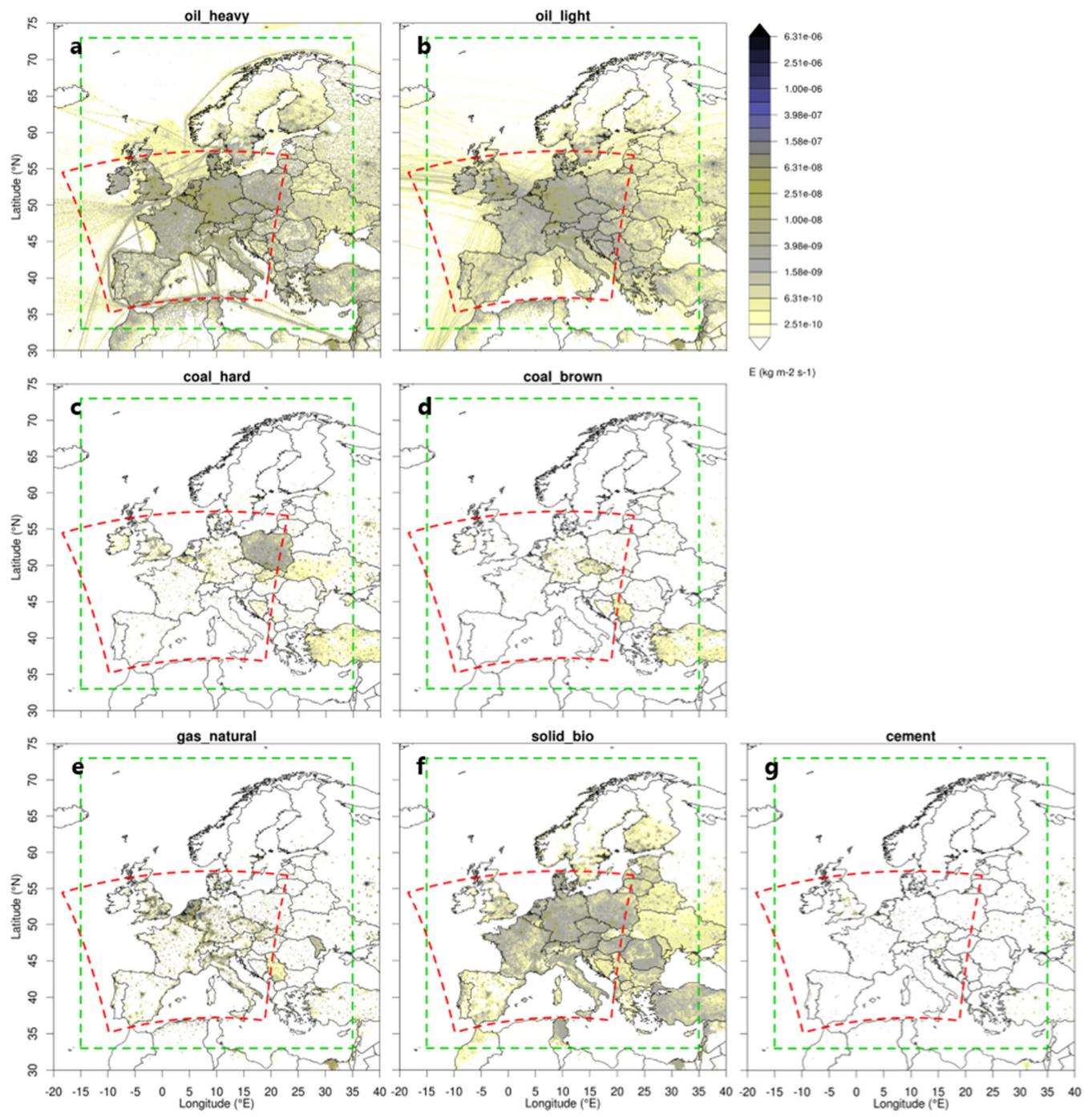
Factor	2009	2010	2011	2012	2013	2014	2015	2016	2017
<b>oil</b>									
Switzerland	1.078	1	0.970	0.988	1.035	0.928	0.942	0.893	0.918
Austria	0.961	1	0.930	0.927	0.940	0.915	0.919	0.937	0.954
France	1.035	1	0.979	0.951	0.939	0.912	0.908	0.902	0.906
Germany	0.993	1	0.969	0.965	0.983	0.958	0.955	0.975	0.996
Italy	1.026	1	0.964	0.901	0.822	0.777	0.812	0.818	0.824
<b>gas</b>									
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
<b>coal</b>									
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
<b>bio</b>									
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.

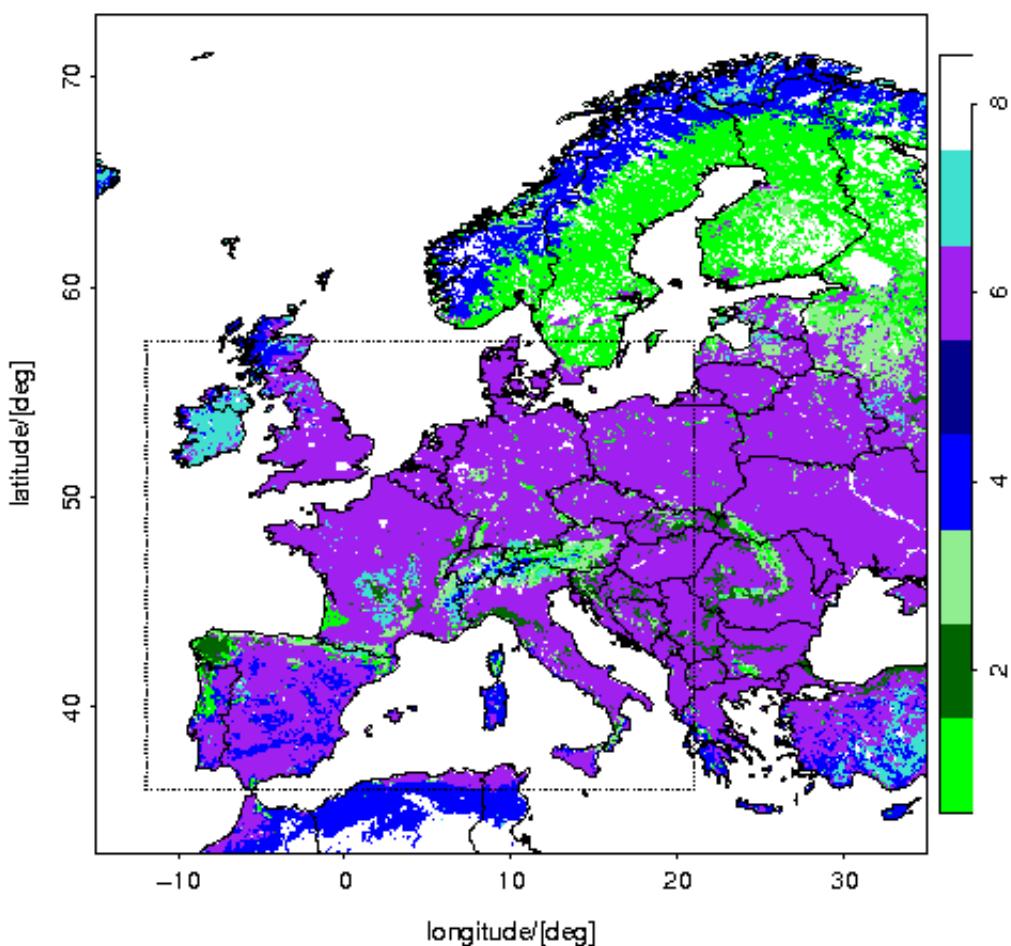
**Table S3.** Time factors to account for seasonal, weekly and diurnal cycles.

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	1.2/1.2/1.2/0.8/0.8/0.8/0.8/0.8/1.2/1.2	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

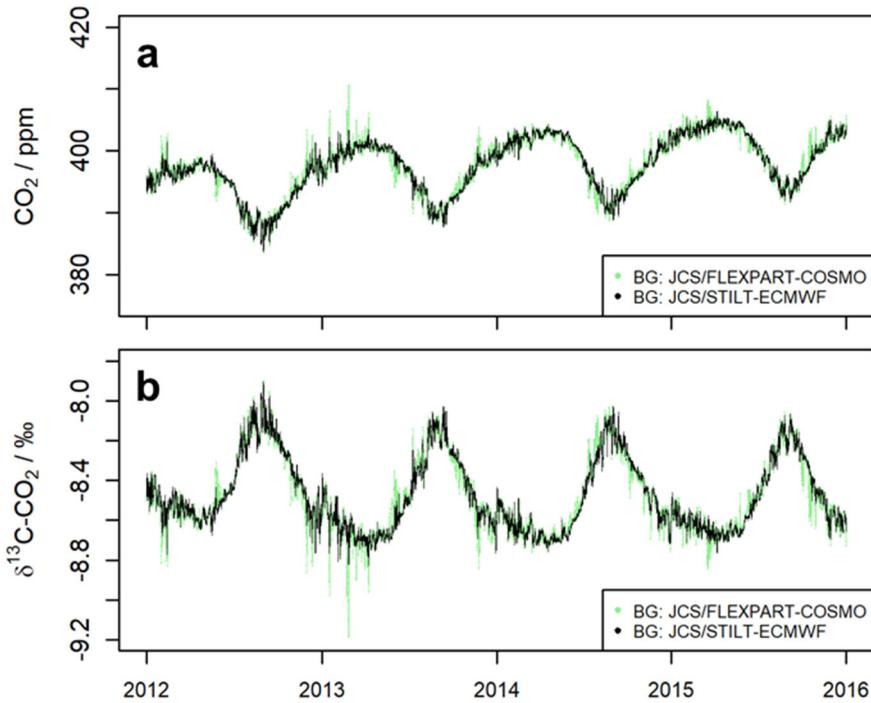
TNO-MACC GNFR Time Factors as of September 2018, e.g. as described in Kuenen et al. (2014).



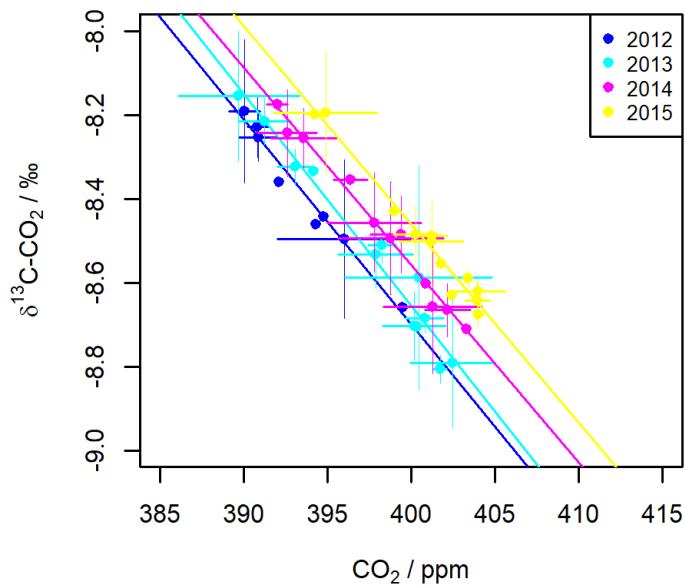
**Figure S1.** EDGARv4.3 anthropogenic emissions maps for the base year 2010 for seven prominent categories concerning CO<sub>2</sub> contributions at JFJ: **a)** oil\_heavy, **b)** oil\_light, **c)** coal\_hard, **d)** coal\_brown, **e)** gas\_natural, **f)** solid biomass, and **g)** cement production, with indication of FLEXPART-COSMO (red) and STILT-ECMWF (green) domain boundaries as used in our study.



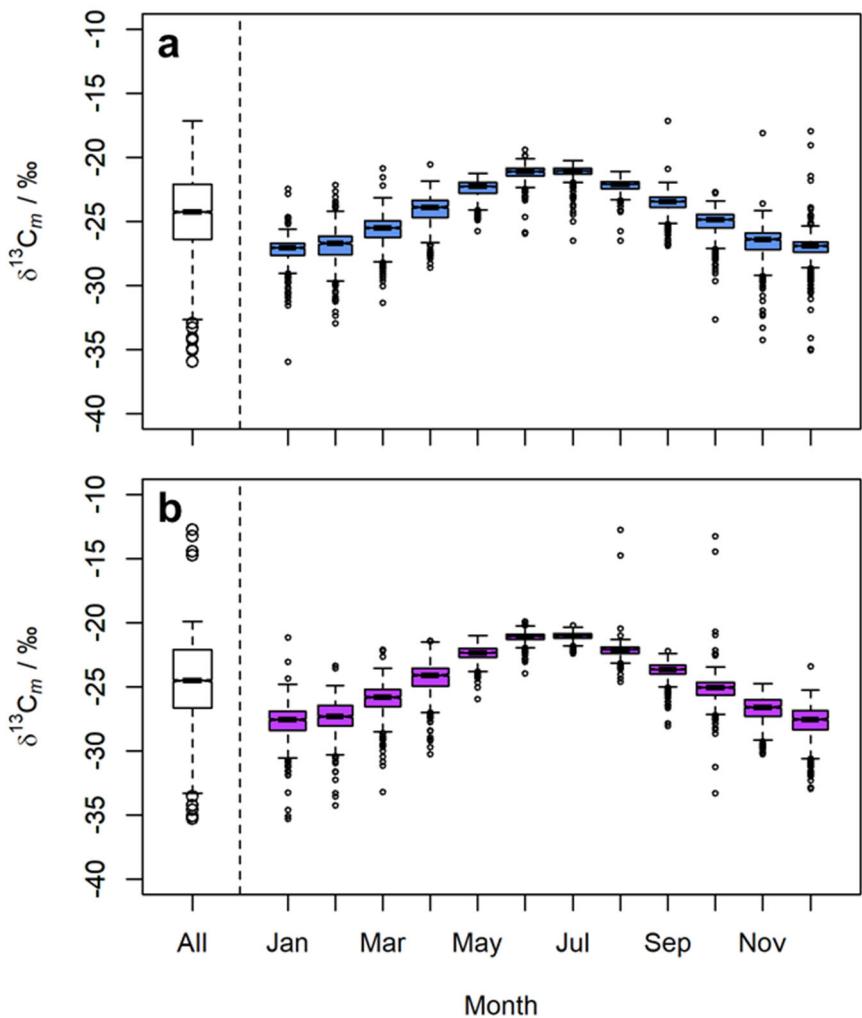
**Figure S2.** Dominant vegetation type (VT) per grid. These VTs include 1) evergreen, 2) deciduous, and 3) mixed forest, 4) shrubland (including open and closed shrubland), 5) savanna (savannas and woody savannas), 6) cropland, 7) grass- land (grassland, cropland/natural vegetation mosaic, and barren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



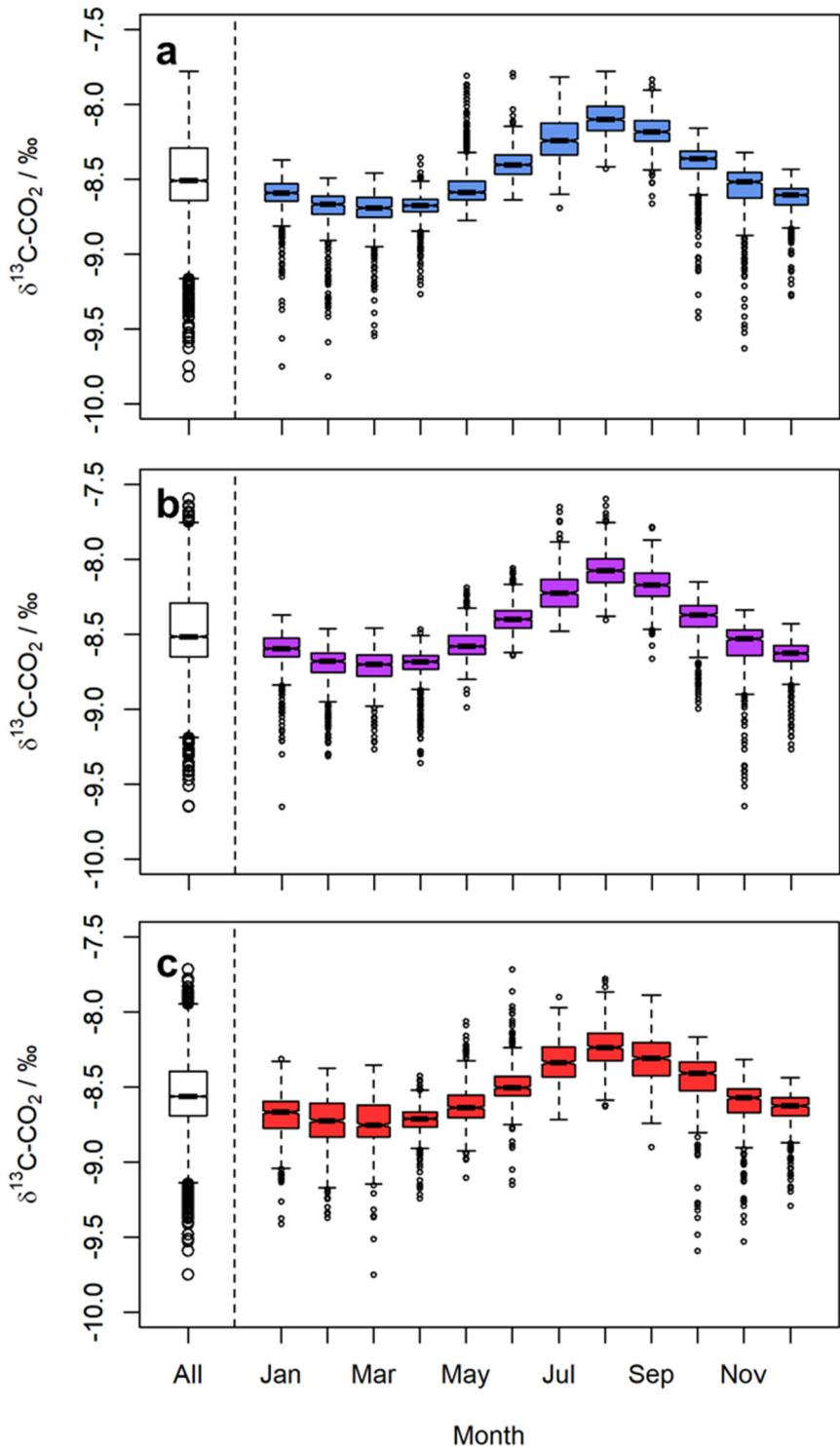
**Figure S3.** Time series of **a)** Jena CarboScope (JCS)  $\text{CO}_2$  concentration background ( $f_b$ ) and **b)**  $\delta^{13}\text{C}-\text{CO}_2$  background (based on  $f_b$  and yearly fits from Figure S4a).



**Figure S4.** Concentration-dependent factors derived from air flask samples (data are provided by MPI-BGC).



**Figure S5** Multi-annual boxplots of model-based mixed source signatures ( $\delta^{13}\text{C}_m$ ) 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  $\delta^{13}\text{C}-\text{CO}_2$  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.

**Table S4.** Results from the CO<sub>2</sub> sensitivity analysis for JJA (2012-2015) and the effect on the regional δ<sup>13</sup>C-CO<sub>2</sub> simulation.

		FLEXPART-COSMO (2012-2015, JJA)						STILT-ECMWF (2012-2015, JJA)				
CO <sub>2</sub> regional	base	sc1 (anthr)	sc2 (nee)	sc3 (gee)	sc4 (resp)	sc5 (trans)	base	sc1 (anthr)	sc2a (nee)	sc3 (gee)	sc4 (resp)	sc5 (trans)
<i>scaling factor, slope</i>		3.14	0.80	0.79	2.08	0.82		3.73	0.72	0.74	1.98	0.74
<i>scaling factor, intercept</i>		0.02	1.04	0.45	-0.88	-0.88		-0.11	1.22	0.49	-0.56	1.54
r <sup>2</sup> (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
δ <sup>13</sup> 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 <sup>2</sup> (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

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