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Seasonal source variability of carbonaceous aerosols at the Rwanda Climate Observatory

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Table S1. Concentrations of carbonaceous components (μgC m^{-3}), inorganic ions (μg m^{-3}) and carbon isotope signatures (‰) of TC in $PM_{2.5}$ at RCO during October 2014 to September 2015. Sampling occurred between 01.00 AM and 06.00 AM C.A.T.

Start	Stop	TC	OC	EC	WSOC	NO_3^-	SO_4^{2-}	NH_4^+	K^+	$\Delta^{14}C_{TC}$	$\delta^{13}C_{TC}$
2014-10-01	2014-10-07	2.76	2.46	0.30	1.90	0.15	0.98	0.37	0.17	-19	-23.6
2014-10-08	2014-10-14	2.80	2.50	0.30	1.55	0.12	0.68	0.23	0.15	N/A	N/A
2014-10-22	2014-10-29	1.58	1.41	0.16	0.93	0.01	0.58	0.19	0.08	-44	-24.0
2014-11-05	2014-11-18	1.03	0.93	0.10	0.56	0.01	0.51	0.15	0.03	-84*	-26.1*
2014-11-19	2014-11-25	1.43	1.28	0.15	1.32	0.01	0.57	0.19	0.04	-84*	-26.1*
2015-04-25	2015-05-01	2.61	2.42	0.19	1.42	0.07	0.80	0.24	0.06	-24	-27.3
2015-05-02	2015-05-08	1.51	1.41	0.11	0.98	0.05	0.29	0.13	0.03	N/A	N/A
2015-05-09	2015-05-15	1.89	1.77	0.12	1.20	0.09	0.27	0.14	0.04	-3	-25.8
2015-05-16	2015-05-22	3.45	3.23	0.22	2.25	0.27	1.02	0.37	0.08	N/A	N/A
2015-05-23	2015-05-29	4.99	4.63	0.36	2.79	0.32	1.22	0.49	0.15	+9	-23
2015-05-30	2015-06-05	4.31	3.99	0.32	2.53	0.19	2.08	0.60	0.22	N/A	N/A
2015-06-06	2015-06-12	4.63	4.16	0.47	3.17	0.67	0.80	0.41	0.27	+18	-21.7
2015-06-13	2015-06-19	9.89	9.06	0.83	6.76	0.61	4.99	1.33	0.78	N/A	N/A
2015-06-20	2015-06-26	10.05	8.67	1.38	6.15	1.30	2.63	1.02	0.72	N/A	N/A
2015-06-27	2015-07-03	11.01	9.24	1.77	7.02	1.76	2.22	1.03	0.81	N/A	N/A
2015-07-04	2015-07-10	9.27	7.93	1.34	5.22	1.27	1.55	0.68	0.65	+25	-21.3
2015-07-11	2015-07-17	11.21	9.29	1.92	6.84	1.64	1.53	0.66	0.83	N/A	N/A
2015-07-18	2015-07-24	13.73	11.77	1.97	8.45	2.12	2.33	1.02	1.02	+30	-22.4
2015-07-25	2015-07-31	17.00	14.84	2.16	9.44	2.48	2.43	1.11	1.18	N/A	N/A
2015-08-01	2015-08-07	12.00	10.24	1.77	6.97	1.81	2.42	0.80	1.01	+26	-22.4
2015-08-08	2015-08-10	8.50	6.82	1.68	4.80	1.59	1.14	0.59	0.26	N/A	N/A
2015-08-11	2015-08-17	7.63	6.37	1.26	4.87	0.91	1.47	0.60	0.59	+19	-22.8
2015-08-19	2015-08-24	12.63	11.52	1.48	7.11	1.46	1.48	0.64	0.68	N/A	N/A
2015-09-01	2015-09-07	3.59	3.24	0.35	2.25	0.17	1.79	0.52	0.31	-25	-23.3
2015-09-08	2015-09-14	7.43	6.44	0.99	4.25	0.53	1.83	0.48	0.48	N/A	N/A

* two filters were combined for isotope analysis.

Table S2. Dual carbon isotope constrained fractions (f, in %) and concentrations of TC (μgC m^{-3}) from the three main sources: C_3-plants, C_4 and fossil at RCO during October 2014 to September 2015 for the 'best scenario'.

Start	Stop	f_{C3}	f_{fossil}	f_{C4}	TC_{C3}	TC_{fossil}	TC_{C4}
2014-10-01	2014-10-07	63±4	6±1	30±4	1.8±0.1	0.18±0.03	0.83±0.10
2014-10-22	2014-10-29	74±5	9±2	17±4	1.1±0.1	0.15±0.03	0.27±0.07
2014-11-05	2014-11-25	81±6	11±3	8±6	1.0±0.1	0.14±0.03	0.10±0.07
2015-04-25	2015-05-01	64±4	7±1	29±4	1.7±0.1	0.17±0.03	0.75±0.10
2015-05-09	2015-05-15	70±4	8±1	22±4	1.3±0.1	0.16±0.03	0.41±0.07
2015-05-23	2015-05-29	58±5	5±1	38±5	2.9±0.2	0.23±0.04	1.9±0.2
2015-06-06	2015-06-12	58±5	5±1	37±5	2.7±0.2	0.22±0.04	1.7±0.2
2015-07-04	2015-07-10	54±6	4±1	42±5	5.0±0.5	0.33±0.10	3.9±0.5
2015-07-18	2015-07-24	53±6	3±1	44±6	7.3±0.8	0.43±0.16	6.0±0.8
2015-08-01	2015-08-07	53±6	3±1	43±6	6.4±0.7	0.39±0.14	5.2±0.7
2015-08-11	2015-08-17	55±5	4±1	41±5	4.2±0.4	0.29±0.08	3.1±0.4
2015-09-01	2015-09-07	61±4	5±1	34±4	2.2±0.1	0.20±0.03	1.2±0.1

Table S3. Dual carbon isotope constrained fractions (f, in %) and concentrations of TC (μgC m^{-3}) from the three main sources: C_3-plants, C_4 and fossil at RCO during October 2014 to September 2015 for the 'maximum C_4 KIE scenario'.

Start	Stop	f_{C3}	f_{fossil}	f_{C4}	TC_{C3}	TC_{fossil}	TC_{C4}
2014-10-01	2014-10-07	54±5	6±1	39±5	1.5±0.1	0.18±0.03	1.1±0.1
2014-10-22	2014-10-29	69±6	9±2	22±6	1.1±0.1	0.15±0.03	0.35±0.09
2014-11-05	2014-11-25	78±8	11±3	10±8	0.96±0.10	0.14±0.03	0.13±0.10
2015-04-25	2015-05-01	55±5	7±1	38±5	1.4±0.1	0.17±0.03	0.99±0.13
2015-05-09	2015-05-15	63±5	8±1	29±5	1.2±0.1	0.16±0.03	0.54±0.09
2015-05-23	2015-05-29	45±6	5±1	50±6	2.2±0.3	0.23±0.04	2.5±0.3
2015-06-06	2015-06-12	46±6	5±1	49±6	2.1±0.3	0.22±0.04	2.3±0.3
2015-07-04	2015-07-10	40±7	4±1	56±7	3.7±0.7	0.33±0.10	5.2±0.7
2015-07-18	2015-07-24	39±8	3±1	58±8	5.3±1.1	0.43±0.16	8.0±1.0
2015-08-01	2015-08-07	39±8	3±1	58±8	4.7±0.9	0.39±0.14	6.9±0.9
2015-08-11	2015-08-17	41±7	4±1	54±7	3.2±0.5	0.29±0.08	4.2±0.5
2015-09-01	2015-09-07	50±6	5±1	45±5	1.8±0.2	0.20±0.03	1.6±0.2

Table S4. Dual carbon isotope constrained fractions (f, in %) and concentrations of TC (μgC m^{-3}) from the three main sources: C_3-plants, C_4 and fossil at RCO during October 2014 to September 2015 for the 'minimum C_4 KIE scenario'.

Start	Stop	f_{C3}	f_{fossil}	f_{C4}	TC_{C3}	TC_{fossil}	TC_{C4}
2014-10-01	2014-10-07	72±3	6±1	23±3	1.9±0.1	0.18±0.03	0.60±0.07
2014-10-22	2014-10-29	78±4	9±2	12±3	1.2±0.1	0.15±0.03	0.20±0.05
2014-11-05	2014-11-25	83±5	11±3	6±5	1.0±0.1	0.14±0.03	0.08±0.06
2015-04-25	2015-05-01	72±3	7±1	21±3	1.9±0.1	0.17±0.03	0.56±0.07
2015-05-09	2015-05-15	76±3	8±1	16±3	1.4±0.1	0.16±0.03	0.31±0.05
2015-05-23	2015-05-29	68±3	5±1	28±3	3.4±0.2	0.23±0.04	1.4±0.2
2015-06-06	2015-06-12	68±3	5±1	27±3	3.1±0.2	0.22±0.04	1.3±0.2
2015-07-04	2015-07-10	65±4	4±1	31±4	6.1±0.4	0.33±0.10	2.9±0.4
2015-07-18	2015-07-24	64±4	3±1	32±4	8.9±0.6	0.43±0.16	4.4±0.6
2015-08-01	2015-08-07	65±4	3±1	32±4	7.8±0.5	0.39±0.14	3.8±0.5
2015-08-11	2015-08-17	66±4	4±1	30±4	5.0±0.3	0.29±0.08	2.3±0.3
2015-09-01	2015-09-07	69±3	5±1	25±3	2.5±0.1	0.20±0.03	0.90±0.11

Table S5. Dual carbon isotope constrained fractions (f, in %) and concentrations of TC (μgC m^{-3}) from the three main sources: C_3-plants, C_4 and fossil at RCO during October 2014 to September 2015 for the ‘depleted fossil scenario’.

Start	Stop	f_{C3}	f_{fossil}	f_{C4}	TC_{C3}	TC_{fossil}	TC_{C4}
2014-10-01	2014-10-07	62±4	6±1	31±4	1.7±0.1	0.18±0.03	0.87±0.10
2014-10-22	2014-10-29	72±5	9±2	18±5	1.1±0.1	0.15±0.03	0.29±0.08
2014-11-05	2014-11-25	79±7	11±3	9±7	1.0±0.1	0.14±0.03	0.12±0.08
2015-04-25	2015-05-01	63±4	7±1	31±4	1.6±0.1	0.17±0.03	0.79±0.10
2015-05-09	2015-05-15	69±4	8±1	23±4	1.3±0.1	0.16±0.03	0.44±0.08
2015-05-23	2015-05-29	56±5	5±1	39±5	2.8±0.2	0.23±0.05	2.0±0.2
2015-06-06	2015-06-12	57±5	5±1	39±5	2.6±0.2	0.22±0.04	1.8±0.2
2015-07-04	2015-07-10	52±6	4±1	44±5	4.9±0.5	0.33±0.10	4.1±0.5
2015-07-18	2015-07-24	51±6	3±1	46±6	7.1±0.8	0.43±0.16	6.2±0.8
2015-08-01	2015-08-07	52±6	3±1	45±6	6.2±0.7	0.39±0.14	5.4±0.7
2015-08-11	2015-08-17	54±5	4±1	43±5	4.1±0.4	0.29±0.08	3.3±0.4
2015-09-01	2015-09-07	59±4	5±1	35±4	2.1±0.2	0.20±0.03	1.2±0.1

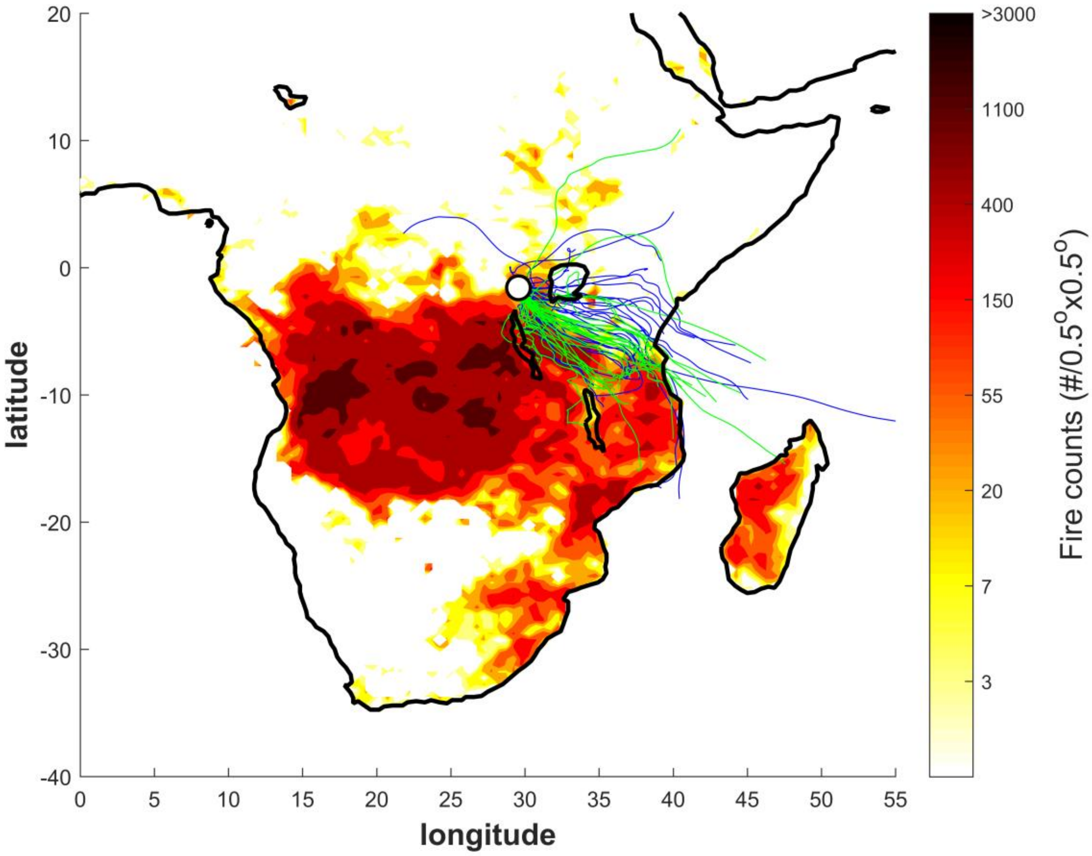

Figure S1 Fire counts and air mass back trajectories for the October 2014 to September 2015 campaign at the Rwanda Climate Observatory (RCO, black and white circle). The fire counts arefrom the Fire Information for Resource Management System (FIRMS) derived from the NASA 659 Moderate Resolution Imaging Spectroradiometer (MODIS) satellite product for June-July-August (JJA), 2015. The thin lines represent daily (4AM, C.A.T.) 5-day air mass back-trajectories arriving at RCO 500 m.a.g.l. (3090 m.a.s.l.). The blue lines correspond to what we here refer to the 'wet' season (October-November 2014 and April-May 2015), whereas the green lines represent the dry JJA season.

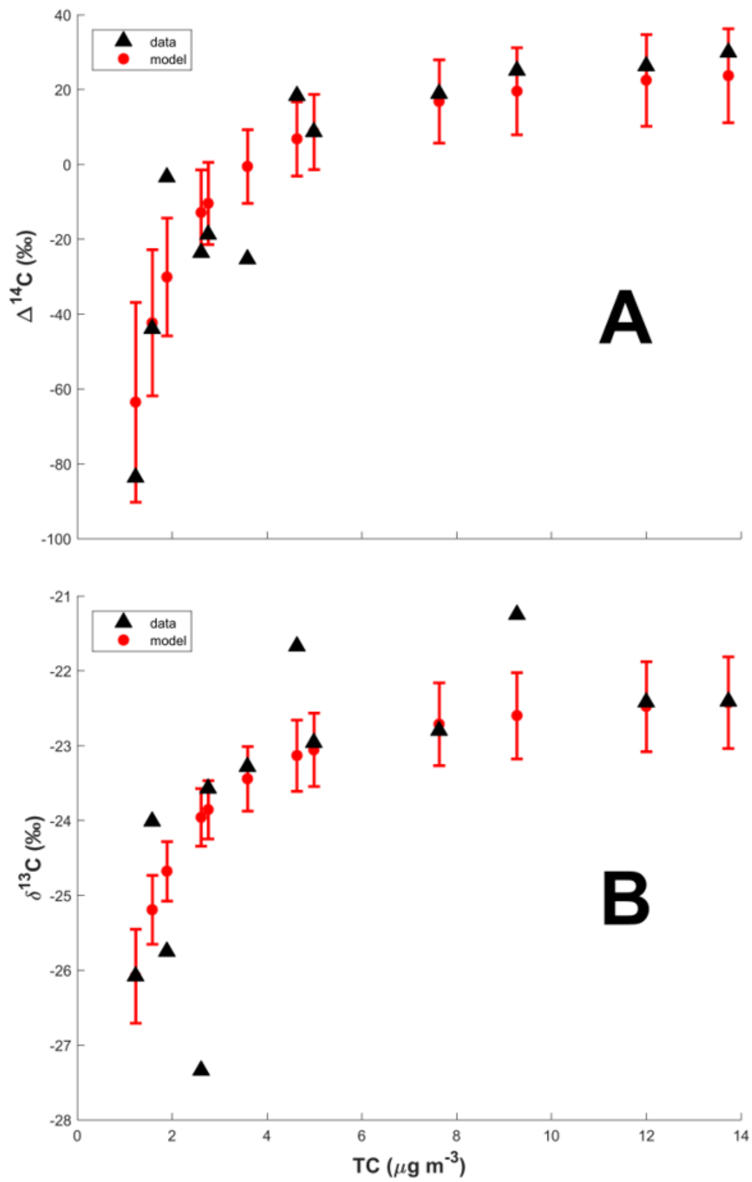

Figure S2. Comparison of observed carbon isotope signatures (black triangles) for TC and values back-calculated from the MCMC-estimated source fractions using the 'best endmember scenario' (red circles with errorbars). Panel A. $\Delta^{14}C$ vs TC. Panel B. $\delta^{13}C$ vs TC.

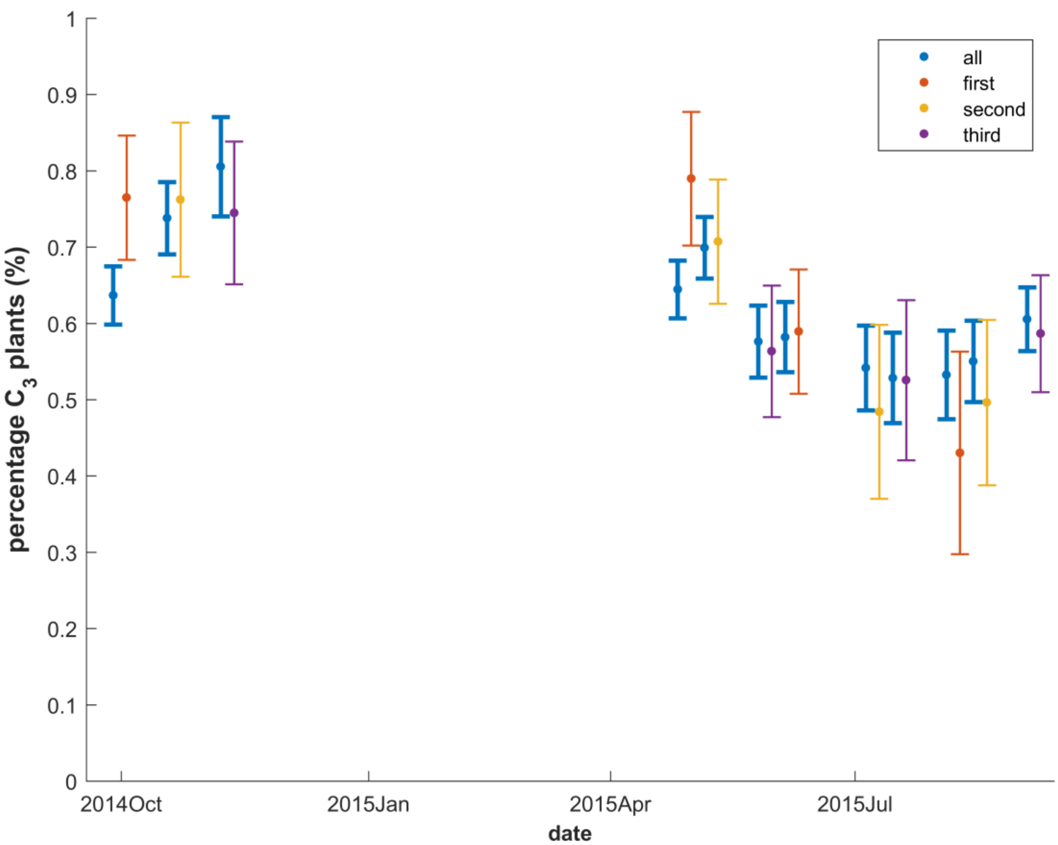

Figure S3 Sensitivity of the Keeling-based Bayesian MCMC source apportionment approach w.r.t. number of data points in the calculation. The fraction C_3-plants is plotted vs time. In blue, the results from using all 12 data pairs (errorbars: mean ± stdev). The orange, yellow and purple lines show calculations using every third data point, starting from data point 1, 2 and 3, respectively. The results from every third data points are shifted slightly in time (to the right) for visual clarity.

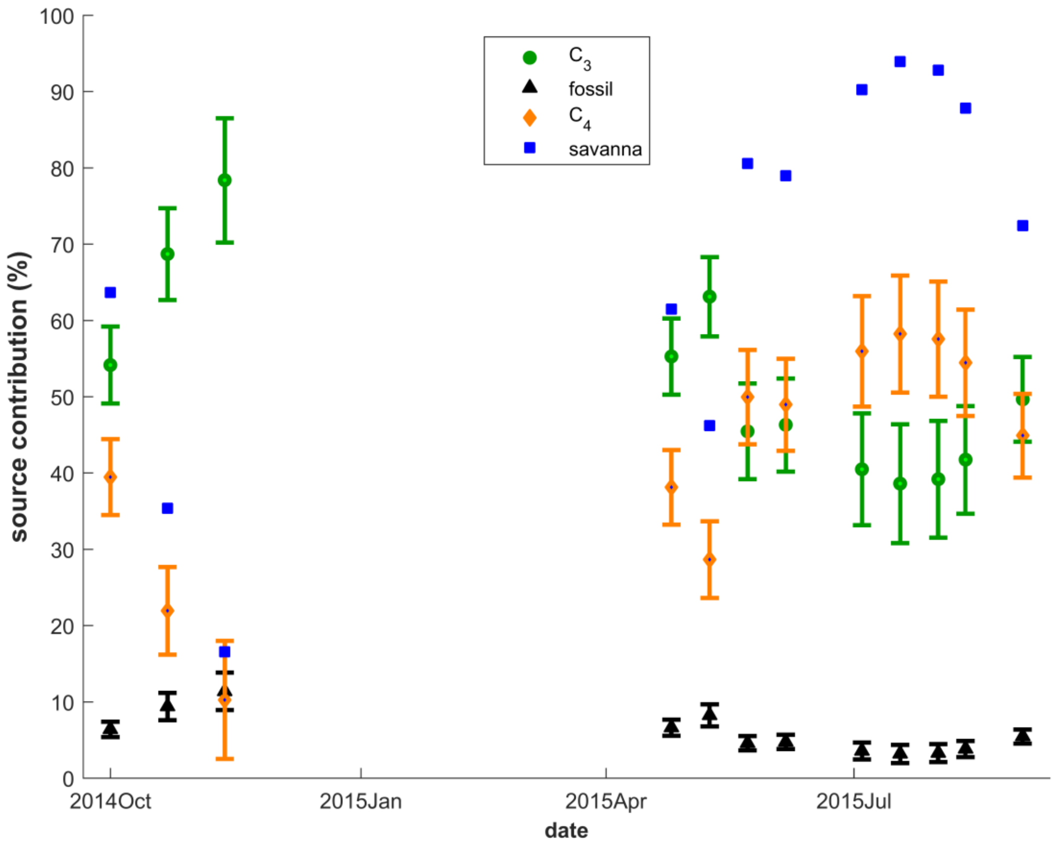

Figure S4: Carbon isotope-source segregated fractions and concentrations of TC vs time computed with the 'maximum C_4 scenario'. Panel A. Relative source contributions (%) of C_3-plants (green circles), C_4-plants (orange diamonds) and fossil (black triangles). Estimated savanna contributions shown as blue squares. The error bars (standard deviations) were constrained using Markov chain Monte Carlo simulations. Panel B. Source segregated concentrations of TC of C_3-plants (green circles), C_4-plants (orange diamonds) and fossil (black triangles).

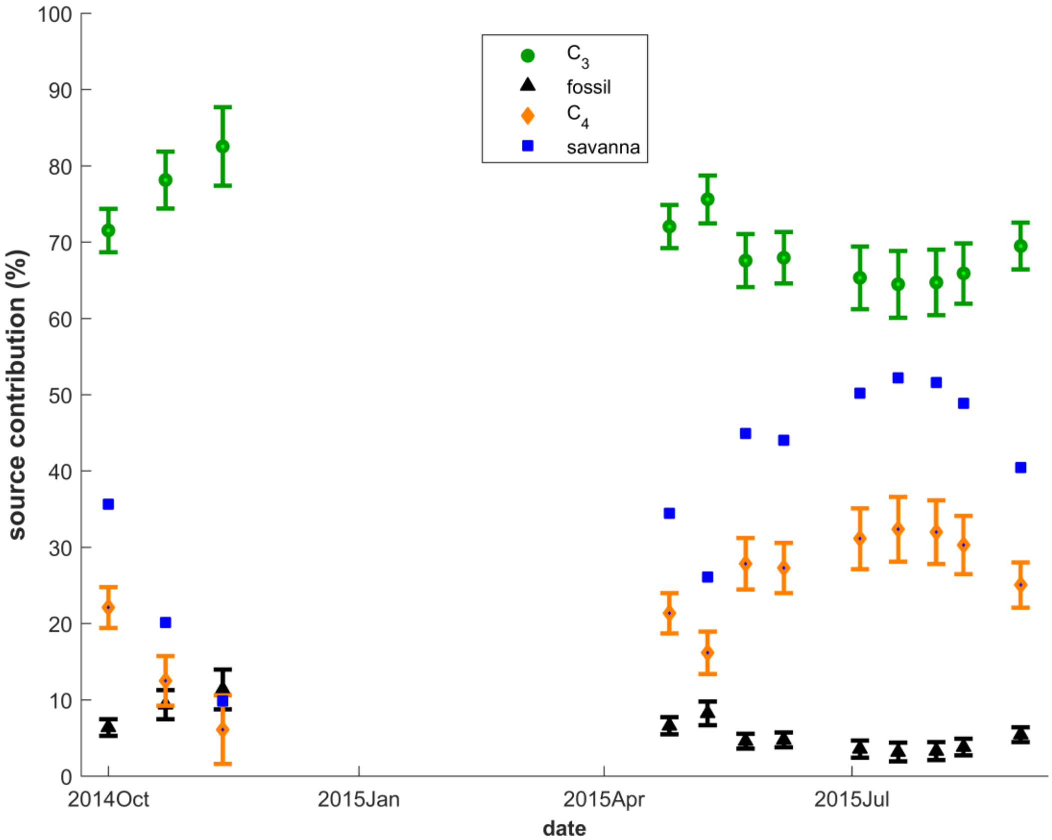

Figure S5: Carbon isotope-source segregated fractions and concentrations of TC vs time computed with the 'minimum C_4 KIE scenario'. Panel A. Relative source contributions (%) of C_3-plants (green circles), C_4-plants (orange diamonds) and fossil (black triangles). Estimated savanna contributions shown as blue squares. The error bars (standard deviations) were constrained using Markov chain Monte Carlo simulations. Panel B. Source segregated concentrations of TC of C_3-plants (green circles), C_4-plants (orange diamonds) and fossil (black triangles).

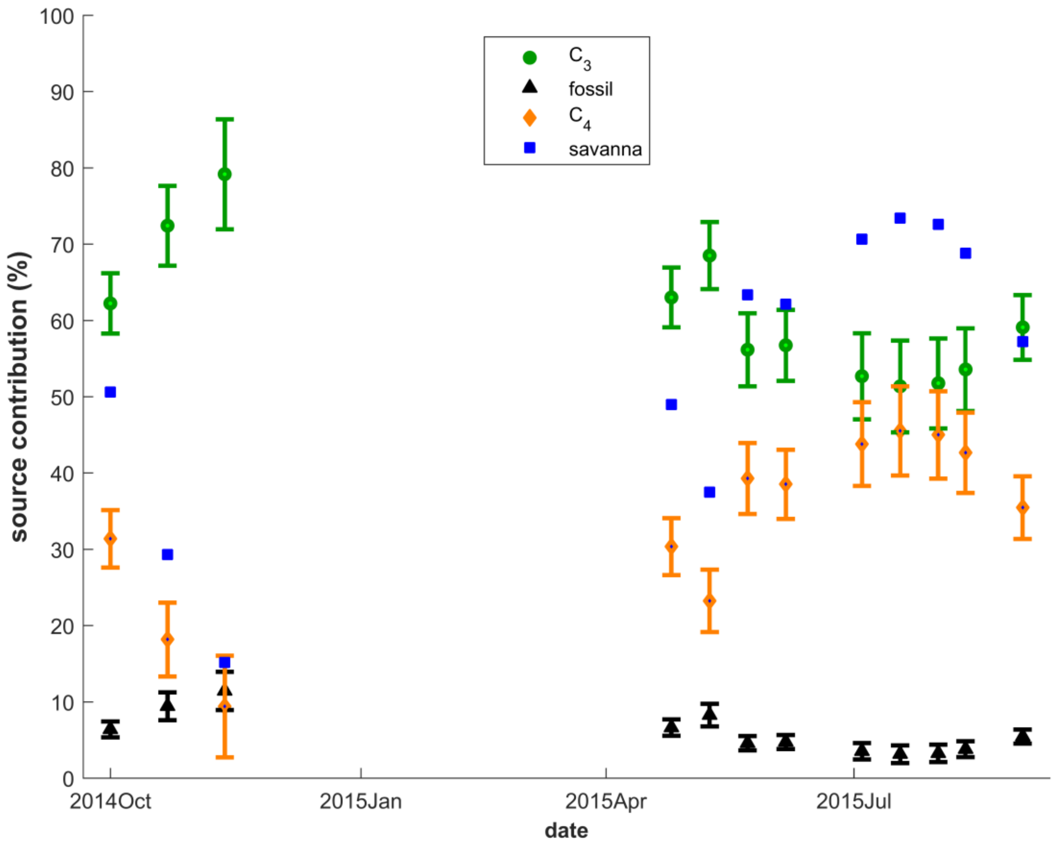

Figure S6: Carbon isotope-source segregated fractions and concentrations of TC vs time computed with the 'depleted fossil scenario'. Panel A. Relative source contributions (%) of C_3-plants (green circles), C_4-plants (orange diamonds) and fossil (black triangles). Estimated savanna contributions shown as blue squares. The error bars (standard deviations) were constrained using Markov chain Monte Carlo simulations. Panel B. Source segregated concentrations of TC of C_3-plants (green circles), C_4-plants (orange diamonds) and fossil (black triangles).