Supplementary Material for: Quantifying and addressing the uncertainties in tropospheric ozone and OH in a global chemistry transport model

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Abstract.

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Table S1. Results from one-at-a-time sensitivity studies showing the average impact of each parameter calculated as "(top_of_range minus bottom_of_range)/two". The metrics considered are the tropospheric O_3 burden, the tropospheric O_3 chemical production and loss rates, the global O_3 deposition rate, the global CH_4 chemical lifetime, and surface O_3 over Europe in summer and winter; the sign indicates the impact of increasing the parameter value.

Rank	Label	Parameter	Scale Factor	O_3 burd (Tg)	O_3 prod (Tg/yr)	O_3 loss (Tg/yr)	O_3 dep (Tg/yr)	CH_4 life (years)	EU O_3 Sum (ppb)	EU O_3 Win (ppb)
Budget terms from model control run:				318.1	4893.6	4542.5	933.4	9.31	50.3	30.4
1	kn2	k(NO_2+OH)	1.60	-31.27	-689.4	-587.3	-101.3	1.47	-7.08	-3.60
2	str	Total O_3 col	20%	27.18	-248.3	-346.0	94.1	1.32	3.67	0.81
3	k1d	k($O(^1D)$+H_2O)	1.30	-23.32	155.6	236.5	-78.2	-0.88	-3.02	-1.17
4	xno	X-sect NO_2	20%	16.35	330.4	269.1	60.5	-0.67	3.46	2.23
5	ddw	Dry dep (water)	3.00	16.31	-92.2	221.0	-315.1	-0.22	5.46	6.35
6	lit	Lightning NO	60%	21.99	328.3	292.4	33.8	-0.68	0.60	1.01
7	iso	Isoprene emis	60%	18.14	480.0	407.9	70.7	0.51	5.26	1.39
8	kch	k(CH_4+OH)	1.15	7.72	129.1	108.1	20.1	-0.89	1.30	0.78
9	knp	k(NO+HO_2)	1.30	13.82	196.4	151.3	43.8	-0.46	2.72	2.05
10	kh2	k(HO_2+O_3)	1.60	-22.12	83.2	158.2	-74.0	-0.04	-3.86	-2.59
11	h2o	Humidity	15%	-14.92	44.7	97.5	-51.2	-0.44	-2.04	-1.00
12	het	Hetero chem	3.00	-10.46	-232.6	-191.1	-40.8	0.45	-1.65	-2.20
13	voc	VOC emission	2.00	9.23	185.9	143.9	40.9	0.18	4.58	4.34
14	knz	k(NO+O_3)	1.20	-9.90	-196.0	-157.7	-37.9	0.39	-2.54	-1.75
15	x1d	X-sect O_3	20%	7.80	-119.3	-148.9	28.3	0.60	1.15	-0.17
16	cnv	Convection	2.00	-14.38	40.7	31.8	18.0	-0.17	0.19	2.18
17	ddg	Dry dep (grass)	3.00	8.78	-15.4	169.2	-184.9	-0.14	3.32	2.63
18	kqn	k($O(^1D)$+N_2)	1.15	8.80	-58.8	-89.4	29.5	0.33	1.14	0.44
19	ddf	Dry dep (forest)	3.00	6.45	-8.4	115.1	-123.6	-0.08	4.00	2.24
20	nox	NO_x emission	25%	5.53	151.6	129.4	21.4	-0.25	1.05	-1.70
21	sno	Soil NO emis	2.00	5.90	169.0	133.8	34.7	-0.29	0.71	0.13
22	kho	k(OH+O_3)	1.45	-4.69	85.0	99.1	-14.3	0.22	-0.73	-0.44
23	pbl	PBL mixing	100.	-4.72	86.3	-45.9	130.4	0.01	-0.84	2.62
24	krh	k(HO_2+RO_2)	1.60	0.97	-108.2	-105.5	-3.1	0.37	-0.38	-0.07
25	aod	Aerosol OD	3.00	1.87	-56.1	-52.1	-4.3	0.21	-0.73	-0.82
26	kn3	k(NO_2+O_3)	1.20	-3.65	-66.8	-51.1	-15.5	0.15	-0.68	-0.96
27	aco	CO emissions	25%	2.71	52.5	44.7	7.1	0.19	0.60	0.39
28	dra	Aerodyn resist	3.00	4.17	32.0	72.8	-40.5	-0.05	1.49	0.88

Table S1. Continued...

Rank	Label	Parameter	Scale Factor	O_3 burd (Tg)	O_3 prod (Tg/yr)	O_3 loss (Tg/yr)	O_3 dep (Tg/yr)	CH_4 life (years)	EU O_3 Sum (ppb)	EU O_3 Win (ppb)
29	opt	Cloud OD	3.00	2.73	47.3	53.8	-4.4	-0.16	-0.57	-0.43
30	kqo	k($O(^1D)$+O_2)	1.15	3.64	-24.3	-37.0	12.2	0.14	0.47	0.18
31	tmp	Temperature	2.0K	-0.31	55.5	52.2	3.1	-0.23	0.24	0.10
32	koh	k(OH+HO_2)	1.30	-0.30	-25.7	-27.2	1.8	0.23	0.08	-0.08
33	fir	Fire emissions	50%	6.48	184.3	154.0	30.2	0.02	0.30	0.26
34	alb	Albedo	30%	-0.43	55.7	53.0	3.0	-0.19	0.29	0.39
35	kni	k(HNO_3+OH)	1.30	2.86	55.8	49.4	7.0	-0.11	0.41	0.17
36	khp	k(OH+H_2O_2)	1.30	0.86	10.4	7.3	3.0	0.20	0.18	0.04
37	xhp	X-sect H_2O_2	20%	-1.16	-8.7	-5.3	-3.2	-0.19	-0.15	-0.05
38	kpd	k(PAN+Δ)	2.00	-2.15	-38.5	-44.3	6.0	0.14	0.34	0.09
39	xna	X-sect $HONO_2$	30%	2.61	52.6	46.8	6.3	-0.12	0.25	0.17
40	ch4	CH_4 burden	4%	2.21	37.0	30.9	5.8	0.12	0.37	0.22
41	kpf	k(NO_2+CH_3CO_2)	1.60	-1.44	12.2	20.2	-7.7	-0.11	-0.30	-0.39
42	krn	k(NO+RO_2)	1.20	1.69	59.7	50.5	9.0	-0.09	0.47	0.33
43	xmp	X-sect CH_3OOH	30%	-0.59	15.5	17.1	-1.4	-0.15	-0.09	-0.00
44	kdn	k(NO_2+NO_3)	1.30	-1.70	-41.8	-34.2	-7.4	0.08	-0.32	-0.30
45	xfm	X-sect HCHO	30%	-0.20	4.9	4.7	0.4	-0.11	0.13	0.44
46	khx	k(H_2+OH)	1.30	0.63	15.5	13.7	1.7	0.10	0.14	0.06
47	air	Aircraft emis	50%	1.70	25.7	22.3	3.3	-0.04	0.15	0.31
48	kpn	k(NO_2+HO_2)	1.30	-1.47	-10.3	-8.0	-2.1	0.02	-0.04	-0.25
49	kco	k(OH+CO)	1.30	0.78	19.2	16.2	3.5	0.01	0.19	0.18
50	koz	k(O_3+Alkene)	1.25	-0.70	-2.1	2.5	-4.5	-0.02	-0.14	-0.09
51	khh	k(HO_2+HO_2)	1.45	-0.10	-104.2	-103.4	-0.5	0.02	-0.28	-0.16
52	eh2	H_2 burden	10%	0.24	5.9	5.2	0.6	0.04	0.05	0.02
53	prc	Precipitation	20%	-0.51	-9.6	-8.8	-1.0	0.02	-0.04	-0.04
54	hna	Henrys Law HNO_3	20%	-0.28	-4.3	-4.0	-0.5	0.01	-0.02	-0.02
55	khn	k(HO_2+NO_3)	1.30	0.20	4.5	3.4	1.1	-0.01	0.01	0.03
56	kis	k(OH+C_5H_8)	1.20	0.05	1.0	-1.2	2.1	-0.01	0.10	0.01
57	hhp	Henrys Law H_2O_2	20%	0.01	-0.1	-0.1	0.0	0.00	0.00	-0.00
58	hfm	Henrys Law HCHO	20%	-0.04	-0.5	-0.5	-0.0	0.00	-0.00	-0.00
59	ko2	k($O(^3P)$+O_2)	1.15	0.01	0.2	0.1	0.0	-0.00	0.00	0.00
60	koo	k(O+O_3)	1.15	-0.00	-0.0	-0.0	-0.0	0.00	-0.00	-0.00

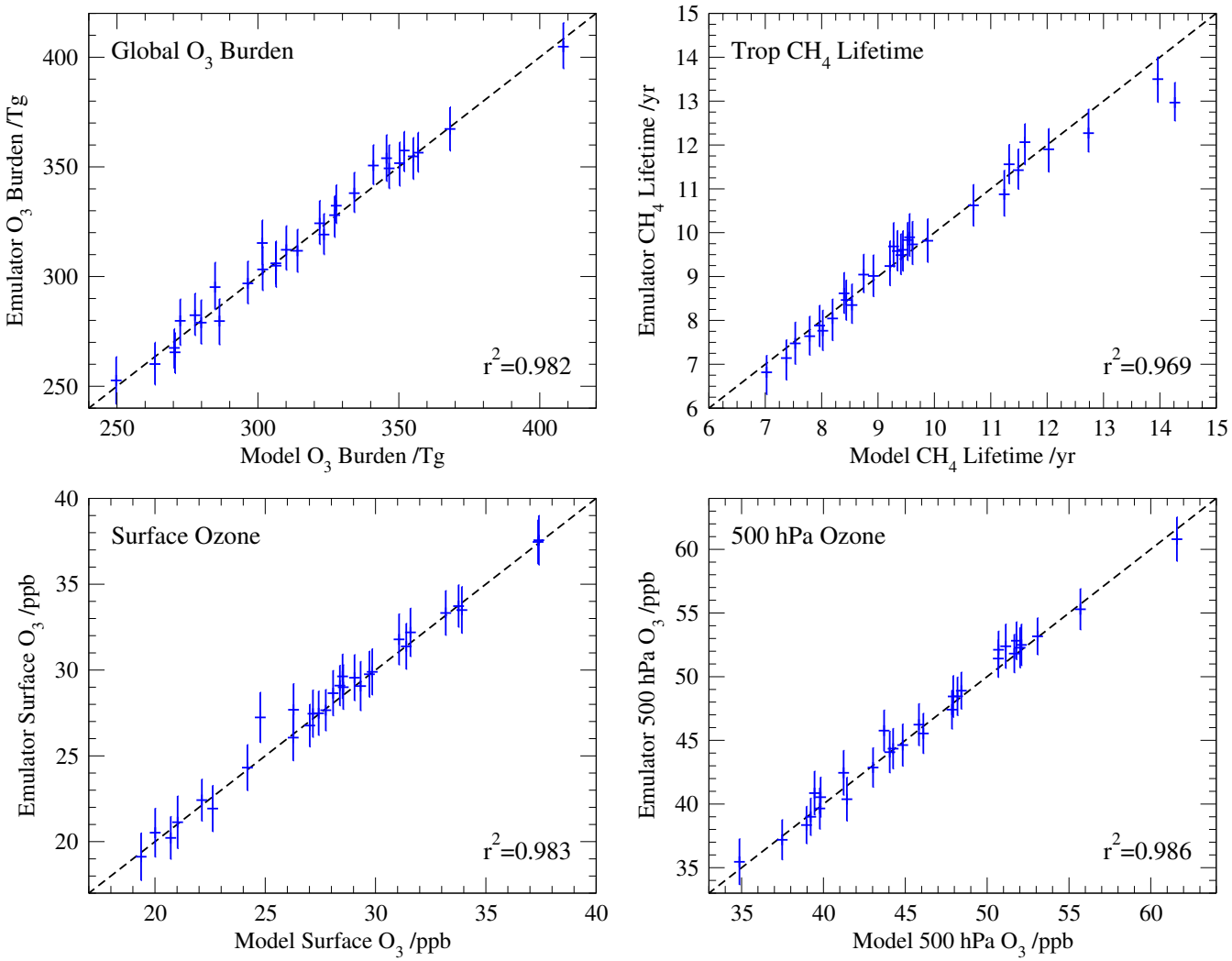

Figure S1. Emulator validation showing the performance of the emulator in reproducing the 30 independent model runs that were not used for training for global tropospheric O_3 burden (a), global CH_4 lifetime (b), global annual mean surface O_3 (c) and global annual mean O_3 at 5 km (d). Error bars show 95% confidence intervals on the emulator predictions.

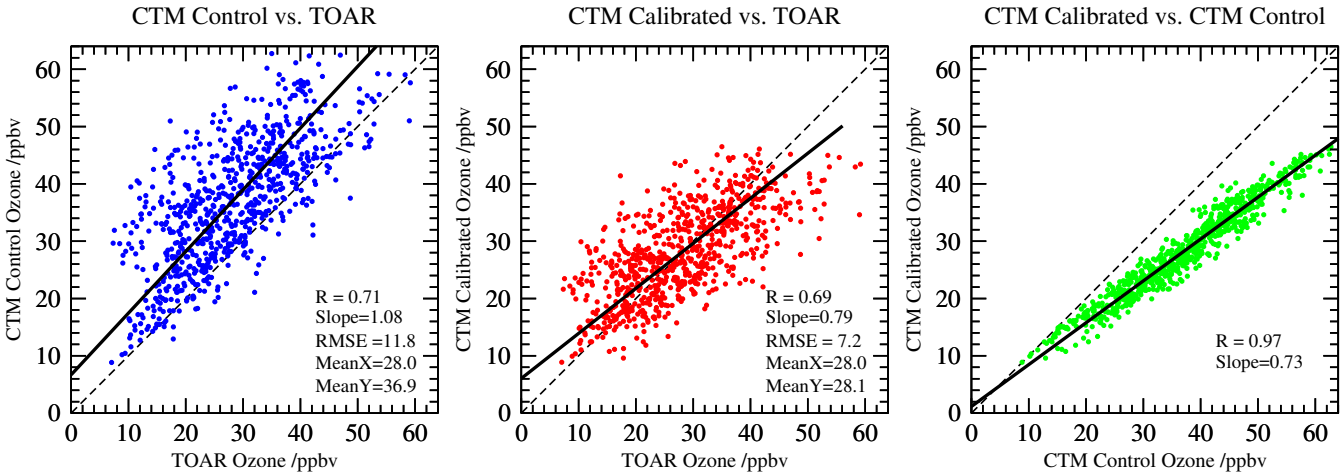

Figure S2. Comparison of model simulated monthly mean surface O_3 at TOAR sites with measurements for the uncalibrated CTM control run (a) and with a calibrated CTM run (b), along with the relationship between calibrated and uncalibrated runs (c).

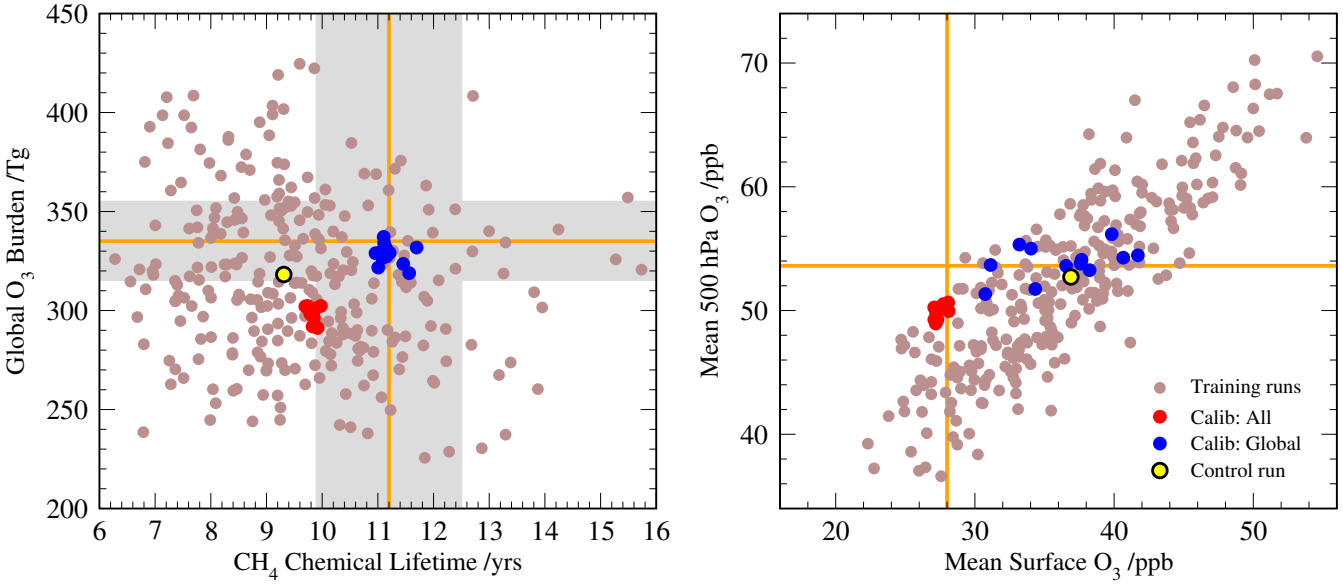

Figure S3. Constraints on model simulated global O_3 and CH_4 metrics (a) and mean surface and 500 hPa O_3 over the measurement sites (b). CTM runs calibrated on all metrics are shown in red, and runs calibrated on the global metrics alone are shown in blue. The uncalibrated (prior) CTM control run is shown in yellow, and CTM runs used for emulator training and validation are shown for context in brown. Mean observation-based values are shown with orange lines, and uncertainty on the global metrics is shown with grey shading.

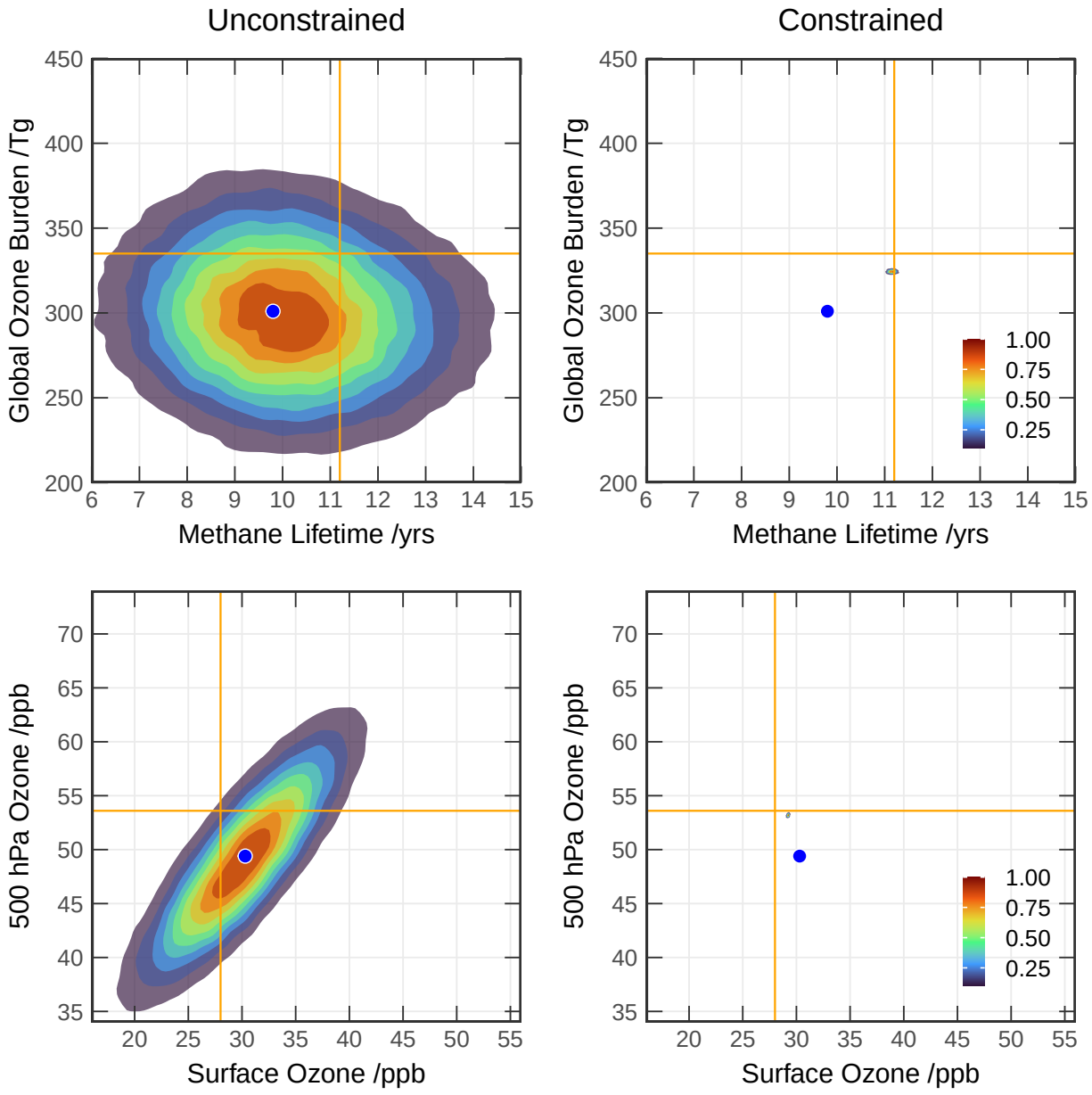

Figure S4. Two-dimensional normalised probability distributions for the global metrics (ozone burden and methane lifetime, upper row) and ozone metrics (500 hPa and surface mixing ratios, lower row) for the sensitivity study exploring the effects of correcting for model structural errors. Panels show the unconstrained (prior) distributions (left column) along with the distributions following constraint (right column). Mean observation-based values are shown with orange lines, and the corrected model control run is shown with a blue circle.

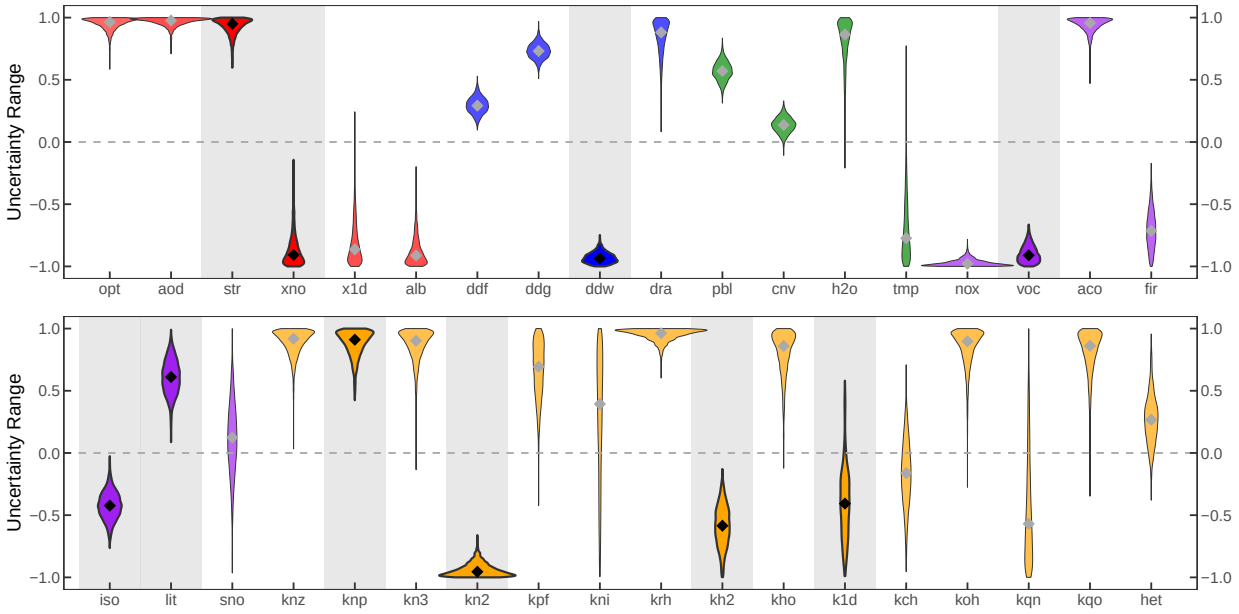

Figure S5. Probability distributions for each parameter following callibration of the model adjusted for structural errors; mean parameter values are shown as points. The 10 parameters with the greatest influence are highlighted in stronger colours on a shaded background.