Supplementary Material: Evidence of a recent decline in UK emissions of HFCs determined by the InTEM inverse model and atmospheric measurements

S1 Measurement Site Information

Table S1 gives details of the instruments used, over which time periods, at each observation site.

Table S1. Instruments at each observation site and when they operated

Site	Operator	Instrument	Years Operated	Sampling Period	Frequency
Mace Head	Uni. of Bristol	GCMS	1994-1998	40 mins	every 6 hr
		GCMS	1998-2003	20 mins	every 4 hr
		GCMS-Medusa	2003-2020	20 mins	every 2 hr
Tacolneston	Uni. of Bristol	GCMS-Medusa	2012-2020	20 mins	every 2 hr
Carnsore Point	Uni. of Bristol	GCMS	2005-2010	20 mins	every 4 hr
Jungfraujoch	Empa	GCMS	2003-2008	20 mins	every 4 hr
		GCMS-Medusa	2008-2020	20 mins	every 2 hr
Monte Cimone	Uni. of Urbino	GCMS	2003-2008	20 mins	every 3 hr
		GCMS	2008-2020	20 mins	every 2 hr
Taunus	Uni. of Frankfurt	GCMS	2013-2020	2 mins	weekly flask

S2 Model versus observation timeseries statistics

Table S2 gives the correlation coefficient between the modelled data and the measurement data per gas per observation site for the InTEM-2yr data for 2018-2019.

Table S2. Correlation coefficient (r) for the match between the observations and modelled time-series at the different sites. These are the results from the 2-yr inversion 2018-2019. NB for CSP used 2008-2009 as no data after 2010

Gas	MHD	TAC	JFJ	CMN	TOB	CSP
HFC-134a	0.97	0.80	0.87	0.74	0.47	0.82
HFC-125	0.96	0.81	0.92	0.80	0.73	0.79
HFC-143a	0.95	0.80	0.79	0.77	0.72	-
HFC-32	0.96	0.79	0.92	0.77	0.25	-
HFC-227ea	0.85	0.21	0.87	0.52	0.16	0.40
HFC-23	0.99	0.93	0.95	0.86	0.32	-
HFC-245fa	0.93	0.55	0.77	0.53	0.52	-
HFC-365mfc	0.87	0.64	0.69	0.60	-	0.71
HFC-152a	0.85	0.84	0.59	0.68	0.64	0.83
HFC-43-10mee	0.54	0.52	0.34	-	-	-

S3 Tabulated results for all HFCs

Table S3. UK emissions 2014 - 2020 estimated using InTEM (1-yr) in Tg CO_2-eq for each HFC and the composite total of all HFCs.

	2014	2015	2016	2017	2018	2019	2020
HFC-134a	4.05±0.44	4.00±0.40	4.43±0.48	4.61±0.55	4.95±0.48	3.20±0.36	2.94±0.33
HFC-125	2.79±0.43	3.42±0.40	3.21±0.40	3.28±0.50	3.38±0.43	2.52±0.33	2.21±0.29
HFC-143a	2.95±0.38	2.95±0.36	2.79±0.41	2.69±0.41	2.13±0.36	1.47±0.25	1.27±0.20
HFC-32	0.17±0.04	0.23±0.03	0.22±0.04	0.28±0.05	0.34±0.07	0.27±0.04	0.25±0.03
HFC-227ea	0.15±0.03	0.17±0.03	0.20±0.04	0.23±0.04	0.25±0.04	0.18±0.03	0.16±0.03
HFC-23	0.21±0.27	0.21±0.26	0.32±0.35	0.25±0.34	0.18±0.24	0.15±0.16	0.11±0.15
HFC-245fa	0.02±0.01	0.04±0.01	0.04±0.01	0.05±0.02	0.05±0.02	0.03±0.01	0.04±0.01
HFC-365mfc	0.09±0.01	0.08±0.01	0.09±0.01	0.06±0.01	0.07±0.01	0.04±0.01	0.04±0.01
HFC-152a	0.01±0.01	0.01±0.01	0.01±0.01	0.02±0.01	0.02±0.01	0.01±0.01	0.01±0.01
HFC-43-10mee	0.01±0.01	0.01±0.01	0.01±0.01	0.01±0.01	0.01±0.01	0.01±0.01	0.01±0.01
Composite	10.46±0.77	11.10±0.72	11.32±0.82	11.48±0.91	11.38±0.78	7.89±0.58	7.04±0.51

Table S4. UK emissions 2008 - 2013 estimated using InTEM (2-yr: 2008 - 2012 & 1-yr: 2013) in Tg CO_2-eq for each HFC and the composite total of all HFCs.

	2008	2009	2010	2011	2012	2013
HFC-134a	3.62±0.29	3.50±0.27	3.86±0.31	4.11±0.35	4.07±0.33	4.30±0.53
HFC-125	2.38±0.19	2.35±0.19	2.66±0.24	3.10±0.28	3.28±0.29	3.62±0.50
HFC-143a	3.25±0.38	3.25±0.36	3.40±0.38	3.45±0.36	3.35±0.30	3.40±0.51
HFC-32	0.14±0.02	0.15±0.02	0.16±0.02	0.17±0.02	0.19±0.02	0.25±0.04
HFC-227ea	0.11±0.02	0.10±0.02	0.12±0.02	0.12±0.03	0.11±0.02	0.11±0.03
HFC-23	0.26±0.28	0.24±0.25	0.29±0.26	0.26±0.23	0.27±0.20	0.26±0.29
HFC-245fa	0.06±0.01	0.05±0.01	0.04±0.01	0.04±0.01	0.03±0.01	0.03±0.01
HFC-365mfc	0.09±0.01	0.06±0.01	0.06±0.01	0.07±0.01	0.06±0.01	0.06±0.01
HFC-152a	0.01±0.01	0.01±0.01	0.02±0.01	0.02±0.01	0.02±0.01	0.01±0.01
HFC-43-10mee				0.01±0.01	0.01±0.01	0.01±0.01
Composite	9.92±0.59	9.70±0.55	10.60±0.61	11.35±0.62	11.39±0.58	12.06±0.94

Table S5. Inventory reported UK emissions of the composite total of all HFCs 2006 - 2018 in Tg CO_2-eq.

2006	2007	2008	2009	2010	2011	2012
14.08±0.90	14.60±0.93	15.12±0.94	15.75±0.95	16.64±1.00	14.93±0.89	15.47±0.90

2013	2014	2015	2016	2017	2018
15.85±0.88	16.04±0.88	15.96±0.85	15.10±0.81	14.00±0.75	12.86±0.68

S4   Results for additional HFCs
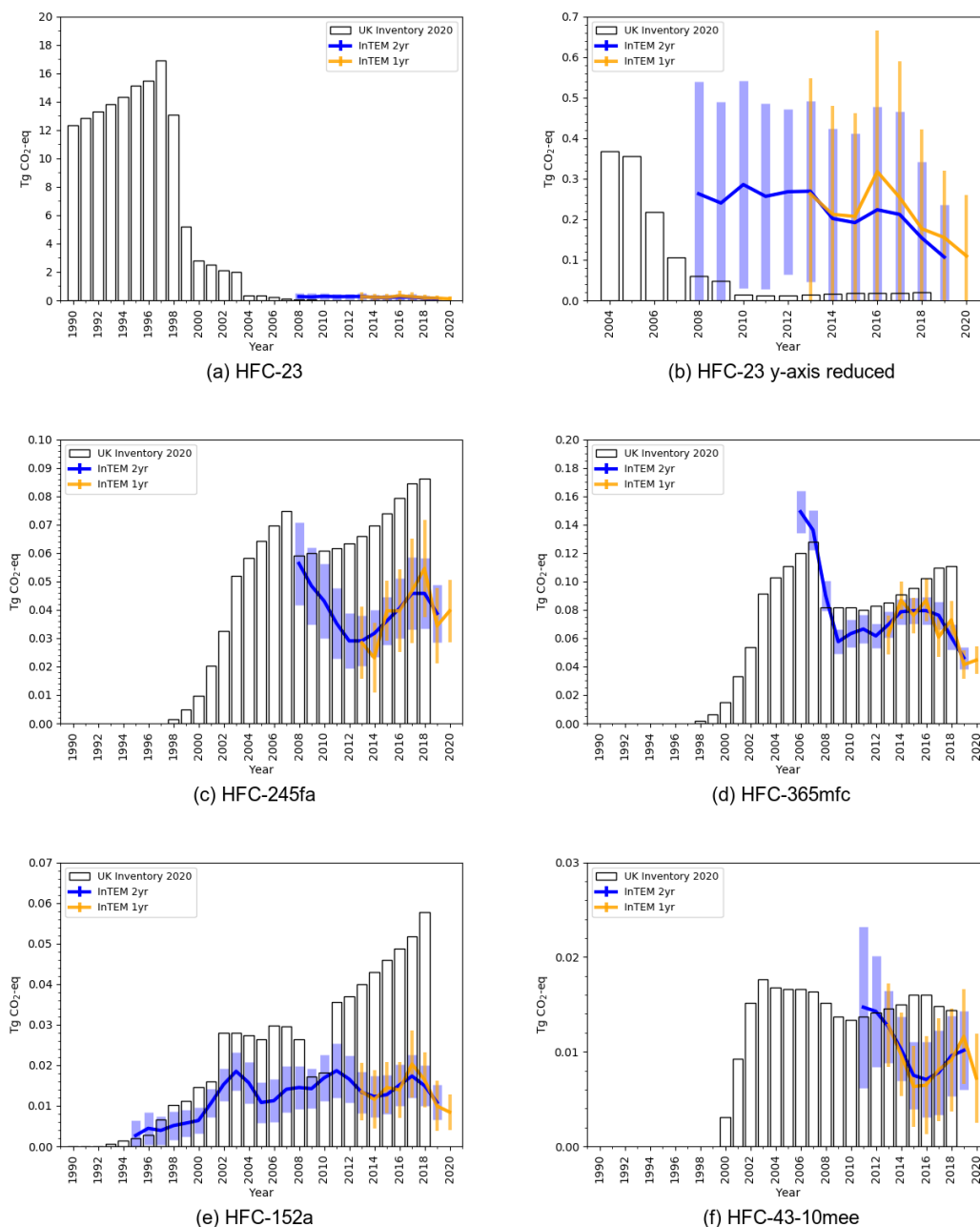
Figure S1. Annual UK emission estimates (Tg CO_2-eq) from the UK 2020 inventory (black), InTEM annualised 2-year inversion (blue) and InTEM 1-year inversion (orange) (a) HFC-23 (b) HFC-23 with y-axis reduced and x-axis sarting in 2004 (c) HFC-245fa (d) HFC-365mfc (e) HFC-152a (f) HFC-43-10mee. The uncertainty bars represent 1 σ.
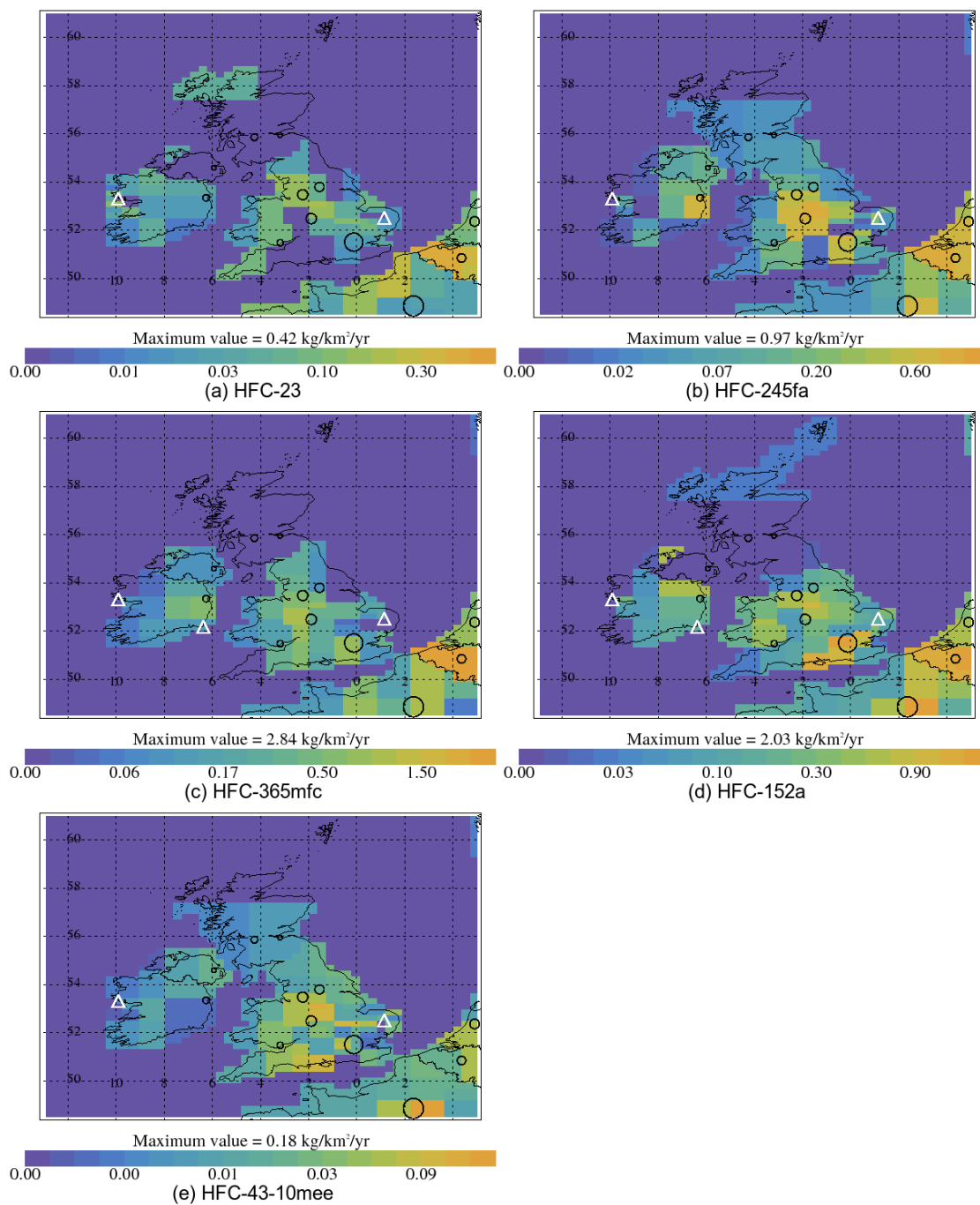
Figure S2. Two-year average InTEM emission estimates 2019–2020 (kg km^{-2} yr^{-1}) (a) HFC-23 (b) HFC-245fa (c) HFC-365mfc (d) HFC-152a (e) HFC-43-10mee. Black circles represent major cities and white triangles show the location of the observation sites.

S4.1   HFC-23

Future emissions of HFC-23, a by-product of HCFC-22 production, are expected to be limited by the Kigali Amendment to the Montreal Protocol, which mandates the destruction of HFC-23 to the extent that is practicable. Future global emission trends of HFC-23 will largely depend on the amount of HCFC-22 produced and the extent to which HFC-23 is destroyed by HCFC-22 production facilities. HCFC-22 is no longer produced in the UK. Fig. S1 shows the UK annual emissions estimated by InTEM plotted with the UK inventory 2020 values. InTEM emission estimates for the UK for HFC-23 from 2009 are higher than the emissions estimated by the inventory for this period, although the InTEM uncertainties are large and mostly extend down to zero. The spatial distribution of HFC-23 emissions over the UK is shown in Fig. S2. The levels of HFC-23 are fairly uniform over the UK. Higher emissions are indicated on the parts of the French coast, Belgium and the Netherlands.

S4.2   HFC-245fa

The InTEM estimates (Fig. S1) have significant uncertainty and are consistently lower than the inventory estimates, with the exception of 2008 and 2009. The inventory estimates show a significant decline from 2007 to 2008 and then a steady annual increase to 2018. The InTEM estimates show a rapid decline from 2008, followed by a rise in emissions from 2014 with a peak in 2018 before a decline from 2018. Fig. S2 shows the spatial InTEM emissions estimate for HFC-245fa over the UK for 2019-2020. HFC-245fa generally follows the distribution of population over the UK. Higher emissions are indicated in Belgium and the Netherlands.

S4.3   HFC-365mfc

The inventory (Fig. S1) shows a sharp decline in emissions in 2008 and the InTEM 2-year estimates show a similar response. Post-2011 the inventory estimates rising UK emissions, a trend initially reproduced by InTEM, however the InTEM estimates then show a sharp decline starting in 2016. The levels of HFC-365mfc emission (Fig. S2 are relatively uniform over England and Wales. Higher emissions are indicated on the near continent in Belgium.

S4.4   HFC-152a

Fig. S1 shows the InTEM and the inventory emission estimates for the UK for HFC-152a for the period 1990 onwards. Between 1999-2008 and from 2011 onwards the inventory estimates are significantly larger than those estimated through inverse modelling. It is also interesting to note the positive trend from 2011 until 2019 in the UK inventory conflicts with a much flatter InTEM trend followed by a sharp decline from 2017 to 2020. The InTEM estimate is approximately one third of the inventory in 2018. Fig. S2 shows the spatial InTEM emissions estimate for HFC-152a over the UK for 2019-2020. The highest emissions are generally focused on the more populated areas, with the highest emission region appearing over London and in the south of the UK.

S4.5   HFC-43-10mee

As estimated by both methods, the UK emissions of this gas are small (Fig. S1. The inventory estimates are initially in agreement with those estimated by InTEM, but the inventory then has a small rise in 2015 and 2016, just as the InTEM emission drops sharply to a low point in 2015 and 2016 before rising back up until 2019 and then dropping sharply in 2020. Throughout, the InTEM uncertainty estimate is large relative to the emission. The spatial distribution is shown in Fig. S2 and is largely distributed by population.