



Supplement of

Country-scale greenhouse gas budgets using shipborne measurements: a case study for the UK and Ireland

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1. Data availability

Table S1: Data availability per season and year of the study. The total number of measured points and the number of points which satisfied the data screening criteria are given.

Year	Season	Total number of points	Number of quality- controlled points	Proportion of quality- controlled points [%]	
2015	Winter	5621	252	4	
2015	Spring	7265	502	7	
2015	Summer	12919	1232	9	
2015	Autumn	7803	493	6	
2015	All seasons	33608	2479	7	
2016	Winter	4198	21	0.5	
2016	Spring	9689	226	2	
2016	Summer	9398	1650	18	
2016	Autumn	20243	1618	8	
2016	All seasons	43528	3515	8	
2017	Winter	3228	618	17	
2017	Spring	14459	1194	8	
2017	Summer	15629	1447	9	
2017	Autumn	3030	272	10	
2017	All seasons	36746	3531	10	
All	Winter	13447	891	6	
All	Spring	31413	1922	6	
All	Summer	37946	4329	11	
All	Autumn	31076	2383	8	
All	All seasons	113882	9525	8	

2. Uncertainty terms

Table S2: Relative contribution of the individual uncertainty terms to the total uncertainty and total uncertainty on the calculated emissions budgets per season and year of the study. The difference between time-lagged and instantaneous emissions budgets illustrates the impact of factoring in the mean West-to-East air mass travel time in the selection of the reference concentrations measured at Mace Head.

		Relative contribution to total uncertainty [%]					Total uncertainty on emissions budget [%]		Difference between time- lagged and instantaneous emissions budgets [%]		
Season	Year	Wind speed in PBL	Molar density	Mole fraction (enhancement above background)	Projection angle θ	Ship speed	CO ₂	CH4	CO ₂	CH₄	Mean air mass travel time ± SD [hour]
Winter	2015	-	-	-	-	-	-	-	-	-	14.7 ± 4.7
Spring	2015	26	4	67	0	2	23	30	2.6	0.9	15.8 ± 5.0
Summer	2015	39	3	54	1	3	288	160	14.5	0.3	23.1 ± 9.9
Autumn	2015	48	5	43	2	2	74	11	2.0	0.3	15.8 ± 5.0
Winter	2016	-	-	-	-	-	-	-	-	-	15.2 ± 0.5
Spring	2016	-	-	-	-	-	-	-	-	-	14.7 ± 2.6
Summer	2016	45	4	49	1	2	63	64	1.4	0.5	20.2 ± 8.8
Autumn	2016	31	3	63	1	2	63	53	0.2	0.2	16.4 ± 7.4
Winter	2017	80	7	8	1	4	5	6	0.4	0.2	13.5 ± 4.1
Spring	2017	62	7	26	1	4	20	29	0.5	0.1	16.4 ± 6.2
Summer	2017	44	4	49	1	2	53	44	2.2	0.2	18.3 ± 6.4
Autumn	2017	71	4	20	1	4	3	4	0.9	0.2	15.9 ± 4.8

3. Planetary Boundary Layer height

Hourly values of the PBL height were derived from the Weather Research and Forecast (WRF) model version 3.7.1 (www.wrf-model.org) (Skamarock et al., 2008) for the spatial domain defined as follows: lower left corner coordinates of 52.0 latitude and -10.0 longitude and the upper right corner of 57.0 latitude and 3.0 longitude. The WRF model initial and boundary conditions were derived from the US National Center for Environmental Prediction (NCEP)/National Center for Atmospheric Research (NCAR) Global Forecast System (GFS) at 1.0°×1.0° resolution (National Centers for Environmental Prediction, 2000), including Newtonian nudging every 6 hours. The Yonsei University Scheme (YSU) planetary boundary layer physics option was used here (Hong et al., 2006).



Figure S1: Daily mean PBL height (solid dots) and standard deviation (shaded ribbon) obtained by averaging the hourly values extracted from the WRF model with YSU scheme for the study period 2015-2017.

4. Seasonality of the prevailing wind direction



Figure S2: Seasonal variability of the prevailing wind direction in the PBL for the three years of the study (2015-2017). The radial unit is the normalised frequency counts of the observations. Plot created with R-package openair (Carslaw and Ropkins, 2012).

References

Carslaw, D.C. and K. Ropkins, (2012) openair — an R package for air quality data analysis. Environ. Modell. Softw., Vol. 27-28, 52-61.