Supplementary Information for:

Quantification of Methane Sources in the Athabasca Oil Sands Region of Alberta by Aircraft Mass-Balance

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Assessment of Uncertainties

Tables S1-6 show the results of the sensitivity analysis to estimate contributions to total uncertainty. Parameters contributing to uncertainties depend on the mass balance method used and the screen-based (Eq. 1) or the box-approach (Eq. 2). Minor uncertainties that contribute to both methods are errors in the CH_4 mixing ratio measurement and wind measurements. CH_4 measurement errors from the instrument are <1%. Measurements of trace species from other instruments were used qualitatively to deduce plume origins, thus they do not contribute to total uncertainties. In a previous study, a Monte Carlo simulation was used to demonstrate the wind measurements contribute <1% to the change in uncertainties (Gordon et al., 2016). A significant source of uncertainty for both mass balance methods is the extrapolation of CH_4 mixing ratios to the surface for ground-level plumes. Surface extrapolation uncertainties are highly variable with flight, consistent with the literature. Cambaliza et al. (2014) found surface extrapolation uncertainties to be 4, 9 and 16% for three different mass balance flights downwind of Indianapolis to determine CH_4 fluxes, and Gordon et al., 2016 found this to be 15% and 26% for two Oil Sands flights for the CNRL facility. The uncertainty depends on the range of surface mixing ratios resulting from fitting varying extrapolation methods. We derive a range of possible emissions rates by comparing results from constant, linear and half-Gaussian extrapolations to the surface. CH_4 measurements at Fort McKay are used as constraints on surface mixing ratios when flight paths are

directly overhead (Aug 16 Flight 4A, SML and SUN). Half-guassian extrapolations are used where fits are above constraints (r^2>0.40). Future studies can further minimize these uncertainties with simultaneous ground-level mixing ratio measurements.

Additional uncertainties specific to the box-approach (Eq. 2) are assessed according to the methodology described in Gordon et al., 2016. Contributing factors are: (1) the uncertainty in the box-top height (affecting the E_{CH} and E_{CV} terms), estimated by reducing the box height by 100 m, (2) changes in air mass density within the volume of the box (affecting E_{CM}), estimated using the minimum and maximum of pressure and temperature ratios derived from surrounding meteorological stations, (3) inclusion of the estimated vertical turbulence term (E_{CVT}), and (4) uncertainty in the mean CH_4 mixing ratio at the box-top (affecting E_{CV}) determined from the 95% confidence interval ($2\sigma/\sqrt{n}$) of interpolated measurements. These terms are recalculated according to the range of possible input parameters in order to derive resulting uncertainties in the emissions rates. Screen-approach specific uncertainties (Eq. 1) are mostly due to the variability in the background mixing ratio $[CH_4]_B$, determined using the outer edges of the screen away from plume sources (screen flights) and upwind measurements (box flights). For each flight measurements from multiple background regions (>1km) occurring closely in time are used as possible inputs, which are identified clearly due to the high CH_4 mixing ratios observed from plumes. Other sources of uncertainty are the vertical extent of the screen (upper bound, z) and the horizontal boundaries (s_1-s_2) of individually characterized plumes. These plume boundaries are expanded and contracted to derive a range of possible integrals.

Uncertainties for each mass balance flight are added in quadrature to derive a range of possible emissions rates. Estimates for the same source category within a facility, as well as total estimates for the same facility, are treated as independent estimates and combined using an error-weighted mean ($1/\sigma^2$).

Meteorological Conditions

Tables S1-6 (bottom) present various flight details and meteorology. Flights used are those with a high number of aircraft transects (≥6) that show full characterization of plume vertical extent. Boundary layer heights are determined using visual inspection of dew point temperature alongside LIDAR backscatter reports from ground-site AMS13 during flight times. Ground temperature and wind direction measurements are based on ground-site data at AMS13 over the course of the day. Wind speeds shown are from interpolated screens ± 1σ.

Table S1-6: **Top: Sensitivity analysis displaying uncertainty contributions (1σ) shown in percent change from the best-estimate emissions rate, added in quadrature for totals. Uncertainties in individual plumes are noted with superscripts for tailings ponds (t), mines (m) and facility/other (f). Screen estimates using an overlapping subset of downwind measurements from a box flight of the same day are shown with an asterisk (*). Middle: List of emissions rates for source categories and facility totals in tonnes CH_4 per hour (tonnes hr^{-1}). Bottom: Various aircraft flight details and meteorological parameters.**

Table S1: Syncrude Mildred Lake (SML)

		Aug 14 Box	Aug 14 Screen A*	Aug 14 Screen B	Aug 16 Screen A
	Measurement Error (%)	1	1	1	1
	Wind Error (%)	1	1	1	1
	Surface Extrapolation (%)	4	11	3	28
Box	Box-top Height (%)	15			
	Density Change (%)	11			
	Vertical Turbulence (%)	2			
	Box-Top Mixing Ratio (%)	**4**			
Screen	Background Mixing Ratio (%)		13	19	8
	Screen-Top Height (%)		6	6	1
	Plume Separation (%)		6^{t}, 11^{m}	5^{t}, 12^{m}	5^{t}, 8^{m}
	Total Uncertainty Facility (%)	20	19	21	30
	Total Uncertainty Plumes (%)		20^{t}, 22^{m}	21^{t}, 24^{m}	30^{t}, 31^{m}
	Emissions Rate Ponds (tonnes hr^{-1})		6.38 ± 1.23	5.83 ± 1.22	8.63 ± 2.59
	Emissions Rate Mines (tonnes hr^{-1})		2.71 ± 0.60	2.67 ± 0.64	3.07 ± 0.95
	Emissions Rate Facility/Other (tonnes hr^{-1})				
	Emissions Rate Total (tonnes hr^{-1})	**7.68 ± 1.54**	**9.10 ± 1.73**	**8.50 ± 1.79**	**11.82 ± 3.55**
	Aircraft Transect Count	6	6	8	9
	Boundary Layer Height (m agl)	360-400	360-400	400-600	350-400
	Temperature (°C)	20.8 ± 6.0	20.8 ± 6.0	20.8 ± 6.0	19.5 ± 3.8
	Wind Speed (m/s)	3.1 ± 2.5	3.1 ± 2.5	5.1 ± 1.6	2.8 ± 0.8
	Daily Mean Wind Direction (°)	220 ± 37	220 ± 37	220 ± 37	225 ± 57

Table S2: Suncor Energy OSG (SUN)

		Aug 16 Screen A	Aug 29 Box	Aug 29 Screen*
	Measurement Error (%)	1	1	1
	Wind Error (%)	1	1	1
	Surface Extrapolation (%)	4	14	4
Box	Box-top Height (%)		1	
	Density Change (%)		17	
	Vertical Turbulence (%)		2	
	Box-Top Mixing Ratio (%)		5	
Screen	Background Mixing Ratio (%)	23		2
	Screen-Top Height (%)	1		9
	Plume Separation (%)	12^{t}, 1^{m}		9^{t}, 9^{m}
	Total Uncertainty Facility (%)	24	23	11
	Total Uncertainty Plumes (%)	27^{t}, 24^{m}		14^{t}, 14^{m}
	Emissions Rate Ponds (tonnes hr^{-1})	3.16 ± 0.85		2.30 ± 0.32
	Emissions Rate Mines (tonnes hr^{-1})	1.53 ± 0.37		1.88 ± 0.26
	Emissions Rate Facility/Other (tonnes hr^{-1})			
	Emissions Rate Total (tonnes hr^{-1})	**4.69 ± 1.13**	**3.96 ± 0.91**	**4.18 ± 0.42**
	Aircraft Transect Count	9	7	7
	Boundary Layer Height (m agl)	350-400	400-500	400-500
	Temperature (°C)	19.5 ± 3.8	15.2 ± 2.4	15.2 ± 2.4
	Wind Speed (m/s)	2.8 ± 0.8	1.8 ± 1.3	1.8 ± 1.3
	Daily Mean Wind Direction (°)	225 ± 57	26 ± 40	26 ± 40

Table S3: Canadian National Resources Limited Horizon (CNRL)

		Aug 20 Box	Aug 20 Screen*	Sep 02 Box	Sep 02 Screen*
	Measurement Error (%)	1	1	1	1
	Wind Error (%)	1	1	1	1
	Surface Extrapolation (%)	22	26	12	11
Box	Box-top Height (%)	1		18	
	Density Change (%)	5		6	
	Vertical Turbulence (%)	2		7	
	Box-Top Mixing Ratio (%)	3		8	
Screen	Background Mixing Ratio (%)		16		25
	Screen-Top Height (%)		5		2
	Plume Separation (%)				6^{m}, 12^{f}
	Total Uncertainty Facility (%)	23	31	25	28
	Total Uncertainty Plumes (%)				29^{m}, 30^{f}
	Emissions Rate Ponds (tonnes hr^{-1})				
	Emissions Rate Mines (tonnes hr^{-1})				2.56 ± 0.74
	Emissions Rate Facility/Other (tonnes hr^{-1})				0.98 ± 0.29
	Emissions Rate Total (tonnes hr^{-1})	**3.65 ± 0.84**	**3.67 ± 1.14**	**3.53 ± 0.88**	**3.54 ± 1.00**
	Aircraft Transect Count	12	12	10	10
	Boundary Layer Height (m agl)	700-900	700-900	600-1000	600-1000
	Temperature (°C)	16.3 ± 4.3	16.3 ± 4.3	12.7 ± 5.1	12.7 ± 5.1
	Wind Speed (m/s)	2.4 ± 1.9	2.4 ± 1.9	5.9 ± 2.8	5.9 ± 2.8
	Daily Mean Wind Direction (°)	262 ± 35	262 ± 35	338 ± 59	338 ± 59

Table S4: Shell Albian and Jackpine (SAJ)

		Aug 21 Box	Aug 21 Screen*	Sep 06 Box	Sep 06 Screen*
	Measurement Error (%)	1	1	1	1
	Wind Error (%)	1	1	1	1
	Surface Extrapolation (%)	5	7	12	7
Box	Box-top Height (%)	8		5	
	Density Change (%)	10		16	
	Vertical Turbulence (%)	5		2	
	Box-Top Mixing Ratio (%)	9		7	
Screen	Background Mixing Ratio (%)		27		17
	Screen-Top Height (%)		10		5
	Plume Separation (%)				
	Total Uncertainty Facility (%)	18	30	22	20
	Total Uncertainty Plumes (%)				
	Emissions Rate Ponds (tonnes hr^{-1})				
	Emissions Rate Mines (tonnes hr^{-1})		1.44 ± 0.43		1.18 ± 0.24
	Emissions Rate Facility/Other (tonnes hr^{-1})				
	Emissions Rate Total (tonnes hr^{-1})	**1.60 ± 0.29**	**1.44 ± 0.43**	**1.25 ± 0.28**	**1.18 ± 0.24**
	Aircraft Transect Count	10	10	10	10
	Boundary Layer Height (m agl)	1200-1500	1200-1500	900-1200	900-1200
	Temperature (°C)	16.5 ± 3.6	16.5 ± 3.6	14.8 ± 6.2	14.8 ± 6.2
	Wind Speed (m/s)	1.3 ± 0.8	1.3 ± 0.8	4.3 ± 0.9	4.3 ± 0.9
	Daily Mean Wind Direction (°)	258 ± 50	258 ± 50	7 ± 50	7 ± 50

Table S5: Syncrude Aurora (SAU)

		Aug 29 Box	Aug 29 Screen*	Sep 06 Screen*
	Measurement Error (%)	1	1	1
	Wind Error (%)	1	1	1
	Surface Extrapolation (%)	10	14	6
Box	Box-top Height (%)	4		
	Density Change (%)	9		
	Vertical Turbulence (%)	2		
	Box-Top Mixing Ratio (%)	3		
Screen	Background Mixing Ratio (%)		11	13
	Screen-Top Height (%)		4	13
	Plume Separation (%)			
	Total Uncertainty Facility (%)	15	19	20
	Total Uncertainty Plumes (%)			
	Emissions Rate Ponds (tonnes hr^{-1})			
	Emissions Rate Mines (tonnes hr^{-1})		1.29 ± 0.25	1.56 ± 0.31
	Emissions Rate Facility/Other (tonnes hr^{-1})			
	Emissions Rate Total (tonnes hr^{-1})	**1.70 ± 0.26**	**1.29 ± 0.25**	**1.56 ± 0.31**
	Aircraft Transect Count	3	3	10
	Boundary Layer Height (m agl)	400-500	400-500	900-1200
	Temperature (°C)	15.2 ± 2.4	15.2 ± 2.4	14.8 ± 6.2
	Wind Speed (m/s)	2.3 ± 0.7	2.3 ± 0.7	4.3 ± 0.9
	Daily Mean Wind Direction (°)	26 ± 40	26 ± 40	7 ± 50

Table S6: Total Oil Sands Screen

		Aug 16 Screen B
	Measurement Error (%)	1
	Wind Error (%)	1
	Surface Extrapolation (%)	3
Box	Box-top Height (%)	
	Density Change (%)	
	Vertical Turbulence (%)	
	Box-Top Mixing Ratio (%)	
Screen	Background Mixing Ratio (%)	14
	Screen-Top Height (%)	5
	Plume Separation (%)	
	Total Uncertainty Facility (%)	16
	Total Uncertainty Plumes (%)	
	Emissions Rate Ponds (tonnes hr^{-1})	
	Emissions Rate Mines (tonnes hr^{-1})	
	Emissions Rate Facility/Other (tonnes hr^{-1})	
	Emissions Rate Total (tonnes hr^{-1})	**23.6± 3.8**
	Aircraft Transect Count	10
	Boundary Layer Height (m agl)	400-450
	Temperature (°C)	19.5 ± 3.8
	Wind Speed (m/s)	2.8 ± 1.0
	Daily Mean Wind Direction (°)	225 ± 57