Electronic Supporting Information for

Seasonal and Latitudinal Variability in the Atmospheric Concentrations of Cyclic Volatile Methyl Siloxanes in the Northern Hemisphere

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Table S1: Sample location coordinates, the sampling times and average air temperature. Temperatures are estimated from climate normals of mean monthly temperature at locations in the vicinity of the sampling sites and do not necessarily reflect the actual conditions during deployment.

Location	Latitude (°N)	Longitude (°W)	Winter Deployment				Spring Deployment				Summer Deployment				Fall Deployment			
			Start	End	Length (d)	Mean Temp. (°C)	Start	End	Lenght (d)	Mean Temp. (°C)	Start	End	Length (d)	Mean Temp. (°C)	Start	End	Length (d)	Mean Temp. (°C)
Canada Transect																		
Alert, Nunavut	82.45	62.51	11/23/18	2/18/19	87	-31	2/18/19	5/22/19	93	-25	5/22/19	9/9/19	110	0	9/9/19	11/28/19	80	-19
Iqaluit, Nunavut	63.79	68.56	Not continued															
Kuujjuaq, Quebec	58.07	68.41	11/27/18	2/18/19	83	-21	2/18/19	5/24/19	95	-12	5/24/19	9/10/19	109	9	9/10/19	11/29/19	80	-1
Fraserdale Ontario	49.875	81.57	11/23/18	2/21/19	90	-14	2/21/19	5/22/19	90	-2	5/22/19	9/5/19	106	15	9/5/19	11/28/19	84	3
Algonquin Ontario	45.59	78.55	11/19/18	2/18/19	91	-10	2/18/19	5/22/19	93	1	5/22/19	9/6/19	107	17	9/6/19	11/28/19	83	5
Borden, Ontario	44.32	79.94	11/19/18	2/19/19	92	-5	2/19/19	5/22/19	92	3	5/22/19	9/6/19	107	18	9/6/19	11/28/19	83	8
Downview, Ontario	43.78	79.47	11/19/18	2/18/19	91	-2	2/18/19	5/22/19	93	6	5/22/19	9/6/19	107	20	9/6/19	11/28/19	83	10
Toronto, Ontario	43.67	79.41	11/16/18	2/17/19	93		2/17/19	5/21/19	93		5/21/19	9/6/19	108		9/6/19	11/28/19	83	
European (EU) Transect																		
Zeppelin, Svalbard	78.9	11.89	1/16/20	4/1/20	76	-13	3/5/19	5/29/19	85	-13	5/29/19	9/18/19	112	3	9/18/19	11/22/19	65	-4
Gortinak, Norway	70.18	28.72	12/1/18	3/3/19	92	-10	3/3/19	6/2/19	91	-4	6/2/19	8/30/19	89	10	8/30/19	12/1/19	93	0
Andøya, Norway	69.14	15.77	11/28/18	2/13/19	77	-1	2/13/19	6/3/19	110	2	6/3/19	9/2/19	91	10	9/2/19	12/5/19	94	5
Tustervatn, Norway	65.83	13.92	12/5/18	3/6/19	91	-2	3/6/19	6/20/19	106	1	6/20/19	9/19/19	91	12	9/19/19	12/5/19	77	3
Kårvatn, Norway	59.87	8.73	1/15/19	3/7/19	51	2	3/7/19	6/3/19	88	5	6/3/19	9/18/19	107	13	9/18/19	12/20/19	93	7
Birkenes, Norway	58.38	8.25	11/28/18	3/2/19	94	1	3/2/19	6/1/19	91	5	6/1/19	9/2/19	93	15	9/2/19	12/7/19	96	8
Blæsbjerg, Denmark	56.3	8.43	12/4/18	3/6/19	92	1	3/6/19	6/4/19	90	5	6/4/19	9/3/19	91	15	9/3/19	12/4/19	92	9
Weilerswist, Germany	50.73	6.84	12/31/18	3/15/19	74	3	3/15/19	6/4/19	81	9	6/4/19	9/15/19	103	17	9/15/19	12/13/19	89	10

Text S1: Standards and Solvents

Three unlabeled cVMS (octamethylcyclotetrasiloxane (D4), decamethylcyclopentasiloxane (D5) and dodecamethylcyclohexasiloxane (D6)) were purchased from Sigma-Aldrich (purity > 99%). Three isotope-labeled cVMS, ^{13}C-D4, ^{13}C-D5 and ^{13}C-D6, were used as internal standards. Those used by the NILU laboratory were purchased from Cambridge Isotope Laboratories with purity all about 99%, while those used by Dow were made in house by Dow Corning Corporation with isotopic purities >98.7% for D4 and > 99% for D5 and D6. All solvents used for equipment cleaning (i.e., acetone and n-hexane) and sorbent extraction (n-hexane) were HPLC grade from Fisher Scientific.

Table S2: Instrument and operation conditions used by Dow. Those used by NILU are provided in Warner et al.[31]

GC Instrument	Agilent 7890A
Mass Detector	Agilent 5975C
GC Column	DB-WAXetr 30m x 250um x 0.25um
Inlet Type	Cup splitter liner
Injection mode	Splitless
Inlet Temperature	150°C
Column flow	Constant pressure 22psi
Oven Temperature Program	Initial temperature 40 °C and hold time 3 min; first ramp 30 °C/min to the maximum temperature of 160 °C and second ramp 45 °C/min to the maximum temperature of 240°C.
Monitoring mode and monitoring ions	Source temperature = 230 °C; Quadrupole temperature = 150 °C; Quantitation SIM: 281, 355, 429m/z (native) and 285, 360, 435m/z (IS) for D4, D5 and D6 respectively.

Text S2: Experiment on Stability of cVMS on Ultraclean Resin

We conducted a test to determine the stability of cVMS on the sorbent used in the passive samplers during extended cold storage. The Ultraclean sorbent (Restek, USA) was used as purchased, with no cleaning or alterations. In a clean hood (40% RH at 22 °C), one jar containing ~200 grams of the sorbent was spiked using 100 µL of a stock solution of D4, D5 and D6 in hexane to achieve nominal concentrations of 100, 200 and 100 ng/g sorbent for D4, D5 and D6, respectively. Another replicate jar was spiked with 100 µL stock solution of D5 in hexane as a single test compound to give a nominal concentration of 1300 ng/g sorbent. The jars were closed immediately and mixed on a jar roller overnight and time zero aliquots (~22 grams) were taken directly from the jars the following morning for Day 1 analysis. The remaining sorbent from each jar was used to fill 4 mesh cylinders, which were stored in four separate closed copper storage tubes in a refrigerator at 5 °C. After 100 and 200 days in storage, two tubes from both spiking batches were removed from storage followed by subsequent extraction and analysis as described for the exposed samples in Section 2.

The concentrations displayed in Figure S1 are averages of duplicates. Although variability can be observed after different storage times, there is no evidence of significant changes in composition or concentration during storage. The observed variation may in part be attributed to incomplete mixing of sorbent material. The negligible amounts of D4 and D6 found in sorbent spiked only with D5 after storage for 200 days, suggest that rearrangement reactions of cVMS are minimal on this sorbent under the tested storage conditions.

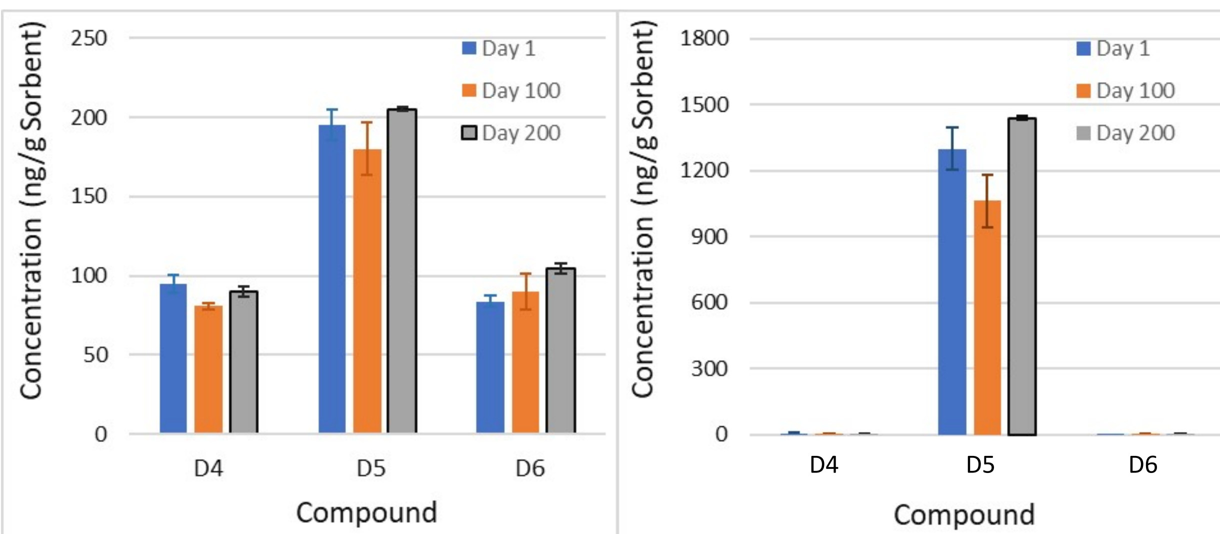

Figure S1: Average concentrations of cVMS measured in Ultraclean resin spiked with a mixture of D4, D5 and D6 (left) or only with D5 (right) after 1, 100 and 200 days in storage at 5 °C. The error bars represent the upper and lower values of the corresponding duplicates analyzed.

Table S3: Concentration in the procedural blanks processed at Dow.

Seasons	Concentration (ng/g Sorb.)		
	D4	D5	D6
Winter	0.22	0.22	0.06
	0.21	0.14	0.07
	0.24	0.12	0.06
Spring	0.32	0.25	
	0.29	0.21	
	0.30	0.18	
Summer/Fall	0.19	0.37	
	0.18	0.13	
	0.17	0.16	
	0.20	0.26	
	0.17	0.14	
	0.20	0.20	
	0.19	0.18	
	0.19	0.12	
	0.21	0.14	

Table S4: Spike recovery obtained at Dow.

Seasons	Spike Recovery		
	D4	D5	D6
Winter	99.7%	105%	104%
	99.7%	99.7%	103%
	98.5%	99.3%	103%
Spring	103%	102%	105%
	107%	106%	108%
	106%	103%	106%
Summer/Fall	106%	107%	106%
	103%	104%	106%
	103%	104%	105%
Average	103%	103%	105%
StDev	3.0%	2.5%	1.5%

Table S5: Concentrations (ng/g sorbent) of D4, D5 and D6 in the field blanks of the Canada transect by two laboratories and their relative difference.

Time	Location	Coordinates		Dow			NILU			Relative Difference (%)		
		Latitude	Longitud	D4	D5	D6	D4	D5	D6	D4	D5	D6
Winter	Alert, Nunavut	82.50	62.33	0.16	4.0	0.20	0.18	2.9	0.050	-10%	31%	120%
	Iqaluit, Nunavut	63.74	68.47	5.4	14	0.37	5.8	14	0.37	-8.5%	-5.2%	0.84%
	Kuujjuaq, Quebec	58.07	68.48	0.63	4.1	0.15	0.45	3.3	0.10	32%	22%	41%
	Fraserdale, Ontario	49.88	81.57	0.48	5.3	0.15	0.21	4.9	0.16	79%	7.9%	-8.2%
	Algonquin, Ontario	45.59	78.55	0.28	4.5	0.073	0.44	3.8	0.14	-44%	16%	-63%
	Borden, Ontario	44.32	79.94	0.28	4.9	0.11	0.38	4.1	0.080	-30%	18%	28%
	Downview, Ontario	43.78	79.47	0.27	4.4	0.060	0.18	2.7	0.059	40%	46%	0.49%
	Toronto, Ontario	43.67	79.41	0.48	4.8	0.017	0.29	4.3	0.36	51%	12%	-180%
Spring	Alert, Nunavut	82.50	62.33	0.15	3.7	0.31	0.13	3.2	0.10	12%	12%	100%
	Kuujjuaq, Quebec	58.07	68.48	1.5	4.1	0.25	1.4	4.0	0.019	1.1%	3.8%	170%
	Fraserdale, Ontario	49.88	81.57	0.66	4.7	0.24	0.27	4.1	0.078	84%	14%	100%
	Algonquin, Ontario	45.59	78.55	0.11	5.4	0.34	0.14	5.3	0.15	-28%	2.2%	78%
	Borden, Ontario	44.32	79.94	0.40	5.0	0.29	0.53	6.1	0.20	-27%	-20%	35%
	Downview, Ontario	43.78	79.47	0.22	5.5	0.41	0.17	5.1	0.074	22%	8.1%	140%
	Toronto, Ontario	43.67	79.41	0.15	4.5	0.33	0.29	8.9	0.96	-66%	-65%	-97%
Summer	Alert, Nunavut	82.50	62.33	0.18	3.6	0.14	0.24	3.6	0.049	-26%	-1.7%	94%
	Kuujjuaq, Quebec	58.07	68.48	3.3	5.2	0.20	0.83	5.3	0.026	120%	-0.61%	150%
	Fraserdale, Ontario	49.88	81.57	0.95	4.3	0.22	0.91	4.4	0.068	5%	-1.8%	110%
	Algonquin, Ontario	45.59	78.55	0.72	6.1	0.60	0.15	4.4	0.13	130%	33%	130%
	Borden, Ontario	44.32	79.94	1.7	6.6	0.19	1.1	6.0	0.16	45%	10%	13%
	Downview, Ontario	43.78	79.47	1.1	6.2	0.23	0.65	6.0	0.24	52%	4.3%	-1.8%
	Toronto, Ontario	43.67	79.41	1.5	4.6	0.22	2.3	5.0	0.081	-46%	-9.0%	93%
Fall	Alert, Nunavut	82.50	62.33	0.24	3.0	0.23	0.14	2.5	0.19	55%	18%	15%
	Kuujjuaq, Quebec	58.07	68.48	3.4	7.9	0.27	3.5	6.4	0.17	-2.4%	20%	46%
	Fraserdale, Ontario	49.88	81.57	1.9	3.3	0.21	1.6	3.6	0.16	16%	-6.5%	25%
	Algonquin, Ontario	45.59	78.55	0.42	11	0.67	0.44	14	0.43	-5.5%	-22%	43%
	Borden, Ontario	44.32	79.94	0.16	9.9	0.56	0.20	8.9	0.45	-19%	10%	21%
	Downview, Ontario	43.78	79.47	0.38	14	0.41	0.39	18	0.70	-1.7%	-21%	-52%
	Toronto, Ontario	43.67	79.41	0.41	9.3	0.38	0.23	5.9	0.84	56%	46%	-76%
Average										17%	6.2%	37%
Stdev										48%	22%	80%

Table S6: Concentrations (ng/g sorbent) of D4, D5 and D6 in the field blanks of the European transect by two laboratories and their relative difference.

Time	Location	Coordinates		Dow			NILU			Relative Difference (%)		
		Latitude	Longitud	D4	D5	D6	D4	D5	D6	D4	D5	D6
Winter	Zeppelin, Svalbard	78.9	11.89	0.19	0.45	0.33	0.22	0.48	0.32	-13%	-7.1%	2.8%
	Gortinak, Norway	70.18	28.72	0.14	0.74	1.9	0.13	0.60	1.7	12%	21%	13%
	Andøya, Norway	69.14	15.77	0.25	1.1	2.9	0.36	1.2	3.7	-39%	-12%	-23%
	Tustervatn, Norway	65.83	13.92	0.12	0.74	1.7	0.17	0.61	1.4	-32%	19%	19%
	Kårvatn, Norway	59.87	8.73	0.11	0.74	1.4	0.12	0.58	1.2	-9.2%	24%	17%
	Birkenes, Norway	58.38	8.25	0.26	1.3	3.2	0.17	0.65	1.8	39%	64%	57%
	Blæsbjerg, Denmark	56.3	8.43	0.092	1.1	1.6	0.31	0.67	1.8	-110%	47%	-15%
	Weilerswist, Germany	50.73	6.84	0.24	2.0	3.7	0.20	0.90	2.7	19%	75%	32%
Spring	Zeppelin, Svalbard	78.9	11.89	0.13	0.82	1.0	0.10	0.33	0.98	32%	85%	5.2%
	Gortinak, Norway	70.18	28.72	0.13	0.51	1.2	0.14	0.36	0.51	-8.6%	35%	81%
	Andøya, Norway	69.14	15.77	0.10	0.45	0.95	0.078	0.23	0.82	28%	66%	14%
	Tustervatn, Norway	65.83	13.92	0.19	0.71	1.3	0.30	0.52	0.43	-47%	31%	99%
	Kårvatn, Norway	59.87	8.73	0.14	1.0	1.2	0.21	0.42	0.94	-41%	85%	25%
	Birkenes, Norway	58.38	8.25	0.24	0.41	1.6	0.23	0.33	1.0	3.7%	21%	44%
	Blæsbjerg, Denmark	56.3	8.43	0.14	0.53	0.99	0.40	1.0	0.66	-96%	-64%	40%
	Weilerswist, Germany	50.73	6.84	0.24	0.67	2.0	-0.018	0.07	0.79	230%	160%	87%
Summer	Zeppelin, Svalbard	78.9	11.89	0.19	0.46	0.72	0.25	0.40	0.74	-27%	14%	-2.3%
	Gortinak, Norway	70.18	28.72	0.25	0.37	0.65	0.37	0.51	0.73	-39%	-33%	-12%
	Andøya, Norway	69.14	15.77	0.27	0.36	0.57	0.25	0.23	0.48	9.4%	45%	16%
	Tustervatn, Norway	65.83	13.92	0.37	0.58	0.61	0.31	0.42	0.58	18%	30%	4%
	Kårvatn, Norway	59.87	8.73	0.25	0.52	0.62	0.23	0.39	0.72	6.0%	28%	-15%
	Birkenes, Norway	58.38	8.25	0.25	0.37	0.62	0.20	0.30	0.47	24%	21%	26%
	Blæsbjerg, Denmark	56.3	8.43	0.25	0.33	0.73	0.27	0.39	0.66	-10%	-17%	11%
	Weilerswist, Germany	50.73	6.84	0.41	0.37	0.66	0.24	0.48	0.25	53%	-26%	90%
Fall	Zeppelin, Svalbard	78.9	11.89	0.15	0.21	0.32	0.16	0.30	0.28	-2.9%	-34%	13%
	Gortinak, Norway	70.18	28.72	1.6	1.6	0.49	0.29	0.34	0.45	140%	130%	7.9%
	Andøya, Norway	69.14	15.77	0.18	0.34	0.41	0.20	0.30	0.39	-14%	12%	3.6%
	Tustervatn, Norway	65.83	13.92	0.31	0.36	0.50	0.27	0.32	0.36	17%	13%	33%
	Kårvatn, Norway	59.87	8.73	0.25	0.31	0.47	0.25	0.33	0.39	-1.2%	-4.5%	20%
	Birkenes, Norway	58.38	8.25	0.14	0.26	0.34	0.22	0.33	0.37	-42%	-22%	-10%
	Blæsbjerg, Denmark	56.3	8.43	0.21	0.36	0.38	0.22	0.33	0.34	-5.2%	10%	11%
	Weilerswist, Germany	50.73	6.84	0.38	0.32	0.56	0.43	0.32	0.45	-13%	0.79%	21%
Average										2.6%	31%	23%
Stdev										60%	47%	31%

Text S3: Field Blank Correction

The amount present in a field blank, expressed as a percentage of the amount present in the exposed sampler from the same deployment site and period, is highly variable between the different cVMS, different sampling sites and the two different sampling transects (Tables S5 and S6). The latter reflects different field blank contamination of the sorbents used for the Canadian and European transects. For example, high percentages observed for D6 in the European transect (~50 % on average) can be attributed to relatively higher D6 levels in the corresponding field blanks. The percentages are small in source regions (e.g., Toronto and Downsview) and higher at remote sites. In extreme cases (e.g., Kuujjuaq), the field blanks contained on average three quarters of the amount found in exposed samples. In only 5 out of 360 samples did levels in field blanks exceed those of the exposed sampler, demonstrating that the applied QA/QC in sampling and analysis yielded high precision and minimal contamination within both field and laboratory.

The percentages tend to be very similar when determined by either Dow or NILU, except for D6 at the remote North American sites, where the blank correction applied by Dow is more than double that applied by NILU. This is the result of D6 field blank levels from Alert, Kuujjuaq and Fraserdale. Although these levels were low, blanks reported by Dow were considerably higher compared to those by NILU.

Table S7: The amount of cVMS in a field blank as a percentage of the amount in the exposed sampler from the same deployment site and period. Shown is the average of four deployment periods.

	Dow			NILU		
	D4	D5	D6	D4	D5	D6
Alert	11%	58%	76%	9%	43%	29%
Kuujjuaq	66%	79%	76%	37%	60%	36%
Fraserdale	21%	35%	49%	16%	35%	29%
Algonquin	7%	32%	51%	4%	35%	31%
Borden	4%	16%	16%	4%	16%	13%
Downsview	1%	4%	3%	0%	4%	4%
Toronto	1%	2%	2%	1%	3%	6%
all of Canada	**16%**	**32%**	**39%**	**10%**	**28%**	**21%**
Zeppelin	5%	7%	56%	5%	6%	60%
Gortinak	18%	9%	72%	11%	6%	72%
Andøya	4%	3%	43%	5%	3%	55%
Tustervatn	10%	8%	65%	9%	6%	56%
Kárvatn	5%	7%	52%	5%	4%	46%
Birkenes	4%	2%	52%	4%	2%	37%
Blæsbjerg	2%	1%	28%	4%	2%	30%
Weilerswist	2%	1%	28%	2%	1%	18%
all of Europe	**6%**	**5%**	**50%**	**5%**	**4%**	**47%**

Table S8: Concentrations (corrected using site- and deployment matched field blanks and normalized both to the mass of sorbent in the sampler and to 90 days of deployment, thus having units of ng per g sorbent per 90 days) of cVMS in passive air samplers deployed during four seasons at different sampling locations along the Canadian transect, measured by Dow and NILU. Bold font: > MQL, Italic font: > MDL, in brackets: < MDL, ND: blank correction gave negative value.

Season	Location	Dow			NILU		
		D4	D5	D6	D4	D5	D6
winter	Alert, Nunavut	*1.6*	*3.6*	(0.034)	**2.5**	*5.3*	*0.22*
	Kuujjuaq, Quebec	*3.0*	*7.4*	*(0.22)*	**4.3**	*8.5*	*0.24*
	Fraserdale, Ontario	**6.4**	**12**	*0.34*	**5.8**	*9.4*	*0.29*
	Algonquin Ontario	**7.4**	**21**	*0.67*	**8.1**	**23**	**0.61**
	Borden, Ontario	**10**	**29**	**1.1**	**8.9**	**24**	**0.92**
	Downsview, Ontario	**72**	**292**	**12**	**57**	**176**	**6.8**
	Toronto, Ontario	**53**	**270**	**12**	**55**	**230**	**9.5**
spring	Alert, Nunavut	*2.8*	*3.4*	(0.12)	**3.9**	*7.2*	*0.37*
	Kuujjuaq, Quebec	*2.5*	*3.7*	(0.088)	**3.8**	*7.4*	*0.21*
	Fraserdale, Ontario	**6.2**	*9.3*	(0.23)	**6.7**	**13**	*0.38*
	Algonquin Ontario	**6.1**	**14**	*0.45*	**6.2**	**14**	*0.62*
	Borden, Ontario	**13**	**33**	**1.5**	**18**	**40**	**1.8**
	Downsview, Ontario	**51**	**128**	**6.2**	**59**	**137**	**6.0**
	Toronto, Ontario	**63**	**299**	**15**	**50**	**149**	**6.7**
summer	Alert, Nunavut	(0.74)	(1.0)	(0.080)	(0.74)	(1.0)	(0.13)
	Kuujjuaq, Quebec	ND	(0.53)	(0.033)	*2.0*	(0.98)	(0.11)
	Fraserdale, Ontario	*2.1*	*4.3*	(0.12)	*2.3*	*4.2*	*0.20*
	Algonquin Ontario	*3.6*	*7.5*	(0.15)	*3.5*	*4.8*	*0.28*
	Borden, Ontario	**13**	**33**	**1.7**	**13**	**20**	**1.1**
	Downsview, Ontario	**68**	**186**	**11**	**76**	**139**	**7.0**
	Toronto, Ontario	**75**	**251**	**14**	**84**	**243**	**14**
fall	Alert, Nunavut	*1.7*	(3.1)	(0.027)	*2.3*	*6.9*	*0.21*
	Kuujjuaq, Quebec	(0.29)	ND	ND	*0.81*	(0.92)	(0.043)
	Fraserdale, Ontario	*3.2*	*7.9*	(0.21)	**4.0**	*7.8*	*0.24*
	Algonquin Ontario	**5.8**	*16*	(0.23)	**6.4**	(13)	(0.31)
	Borden, Ontario	**17**	**40**	**1.5**	**22**	**48**	**1.8**
	Downsview, Ontario	**69**	**239**	**9.6**	**84**	**200**	**7.5**
	Toronto, Ontario	**48**	**216**	**9.7**	**80**	**322**	**14**

Table S9: Concentrations (corrected using site- and deployment matched field blanks and normalized both to the mass of sorbent in the sampler and to 90 days of deployment, thus having units of ng per g sorbent per 90 days) of cVMS in passive air samplers deployed during four seasons at different sampling locations along a European transect, measured by Dow and NILU. Bold font: > MQL, Italic font: > MDL, in brackets: < MDL, ND: blank correction gave negative value.

Season	Location	Dow			NILU		
		D4	D5	D6	D4	D5	D6
winter	Zeppelin, Svalbard*	**10**	**26**	(1.2)	**7.6**	**20**	(0.91)
	Gortinak, Norway	**12**	**21**	(0.27)	**11**	**16**	(0.62)
	Andøya, Norway	**12**	**41**	*3.7*	**12**	**38**	(0.89)
	Tustervatn, Norway	**8.1**	**21**	(0.90)	**7.0**	**17**	(0.80)
	Kårvatn, Norway	**8.3**	**23**	(1.7)	**8.4**	**25**	(2.2)
	Birkenes, Norway	**8.4**	**31**	(0.64)	**8.5**	**32**	(1.9)
	Blæsbjerg, Denmark	**18**	**90**	*4.3*	**11**	**50**	(1.6)
	Weilerswist, Germany	**17**	**129**	*5.5*	**17**	**130**	*5.3*
spring	Zeppelin, Svalbard	**6.1**	**11**	(0.41)	**6.5**	**11**	(0.16)
	Gortinak, Norway	**6.2**	**12**	(0.37)	**6.8**	**16**	(0.54)
	Andøya, Norway	**11**	**25**	*1.3*	**8.6**	**20**	*0.69*
	Tustervatn, Norway	**3.3**	**6.1**	ND	**4.4**	**10**	*0.68*
	Kårvatn, Norway	**3.7**	**6.9**	(0.41)	**5.4**	**15**	*0.94*
	Birkenes, Norway	**6.3**	**20**	(1.0)	**6.0**	**21**	*1.3*
	Blæsbjerg, Denmark	**8.7**	**29**	*1.4*	**5.7**	**32**	**2.4**
	Weilerswist, Germany	**14**	**69**	**3.7**	**10**	**49**	**4.2**
summer	Zeppelin, Svalbard	**1.3**	**2.0**	(0.051)	**1.4**	**1.8**	(0.083)
	Gortinak, Norway	**0.78**	**2.5**	*0.34*	**0.7**	**2.5**	ND
	Andøya, Norway	**2.0**	**7.1**	*0.40*	**1.8**	**4.9**	(0.32)
	Tustervatn, Norway	**1.0**	**3.2**	**0.78**	**0.9**	**2.4**	(0.034)
	Kårvatn, Norway	**2.0**	**6.9**	**0.65**	**2.0**	**5.2**	(0.43)
	Birkenes, Norway	**3.2**	**13**	**0.81**	**3.6**	**12**	*0.94*
	Blæsbjerg, Denmark	**6.0**	**27**	**1.7**	**6.2**	**21**	*1.4*
	Weilerswist, Germany	**14**	**64**	**3.6**	**18**	**77**	**4.7**
fall	Zeppelin, Svalbard	**5.0**	**16**	*0.77*	**5.3**	**15**	**0.70**
	Gortinak, Norway	**2.0**	**9**	*0.35*	**4.2**	**11**	**0.59**
	Andøya, Norway	**7.1**	**22**	**1.0**	**7.2**	**20**	**0.87**
	Tustervatn, Norway	**4.4**	**15**	*0.69*	**5.6**	**17**	**0.88**
	Kårvatn, Norway	**5.3**	**19**	**1.3**	**4.7**	**14**	**0.98**
	Birkenes, Norway	**5.0**	**20**	**1.1**	**5.2**	**20**	**1.2**
	Blæsbjerg, Denmark	**8.6**	**39**	**2.0**	**9.0**	**40**	**2.2**
	Weilerswist, Germany	**12**	**59**	**2.7**	**11**	**64**	**3.3**

*: Winter sampler deployment was not successful at Zeppelin in 2018. The data shown here were from a deployment between January 16 to April 1, 2020

Text S4: Detection Limits

Method detection limit (MDL) and quantitation limit (MQL) were defined as 3 and 10 times the standard deviation of field blanks belonging to a particular sorbent batch. Also, MDL and MQL were calculated separately for samples analyzed by NILU and Dow. In the case of the European transect there was one resin batch for each seasonal deployment. Therefore, the eight sets of MDLs and MQLs for each cVMS were based on the standard deviation of field blanks from eight sites analyzed by one of the labs. For the Canadian transect, the first batch of sorbent was used for all four deployments at the three northern sites as well as the first deployment of the four Southern sites, so that the MDL and MQL were derived from the standard deviation of 16 field blanks. The MDL and MQL for the remaining three deployments at the Southern four locations were based on the standard deviation of four field blanks. Although field blank concentrations belonging to a particular batch were similar, individual outliers were observed in field blanks from certain sampling sites and deployments (Figure 3). These outliers, identified by a ROUT test (false detection rate (Q) = 1%), were excluded from the calculation of MDLs and MQLs (Table S10). This only applied to 10 out of 360 field blank concentrations, with fewer incidences in Europe (2/192) than in Canada (8/168) and for D5 (1/120) and D6 (2/120) than for D4 (7/120). Six of the outliers occurred in the large batch of 16 field blanks from Canada (6/96).

Table S10: MDL and MQL for all samples.

Item	Canada Transect							European Transect						
	Batch	Dow			NILU			Batch	Dow			NILU		
		D4	D5	D6	D4	D5	D6		D4	D5	D6	D4	D5	D6
Stdev of Field Blanks without outliers	big batch*	0.38	1.1	0.080	0.25	1.0	0.060	winter	0.070	0.47	1.1	0.090	0.24	0.71
	spring batch	0.13	0.45	0.050	0.17	1.8	0.070	spring	0.050	0.21	0.36	0.12	0.28	0.22
	summer batch	0.41	0.90	0.19	0.45	0.78	0.070	summer	0.070	0.090	0.060	0.050	0.090	0.17
	fall batch	0.12	2.3	0.13	0.12	5.3	0.20	fall	0.090	0.060	0.080	0.080	0.020	0.060
MDL in ng/g (90 days)	big batch	1.1	3.3	0.25	0.75	3.0	0.17	winter	0.20	1.4	3.3	0.26	0.71	2.12
	spring batch	0.39	1.4	0.16	0.52	5.3	0.20	spring	0.16	0.64	1.1	0.35	0.84	0.67
	summer batch	1.2	2.7	0.58	1.4	2.3	0.20	summer	0.22	0.27	0.17	0.16	0.28	0.51
	fall batch	0.36	7.0	0.40	0.35	16	0.60	fall	0.26	0.17	0.25	0.25	0.05	0.17
MDL in ng/m^3	big batch	0.28	0.82	0.060	0.18	0.74	0.04	winter	0.050	0.35	0.81	0.060	0.18	0.52
	spring batch	0.10	0.33	0.040	0.13	1.3	0.050	spring	0.040	0.16	0.27	0.090	0.21	0.16
	summer batch	0.30	0.66	0.14	0.33	0.58	0.050	summer	0.050	0.070	0.040	0.040	0.070	0.13
	fall batch	0.090	1.7	0.10	0.090	3.9	0.15	fall	0.070	0.040	0.060	0.060	0.010	0.040
MQL in ng/g (90 days)	big batch	3.8	11	0.82	2.5	10	0.56	winter	0.67	4.7	11	0.87	2.4	7.1
	spring batch	1.3	4.5	0.52	1.7	18	0.65	spring	0.52	2.1	3.6	1.2	2.8	2.2
	summer batch	4.1	9.0	2.0	4.5	7.8	0.66	summer	0.74	0.89	0.58	0.52	0.93	1.7
	fall batch	1.2	23	1.3	1.2	53	2.0	fall	0.88	0.55	0.85	0.83	0.16	0.56
MQL in ng/m^3	big batch	0.93	2.7	0.20	0.62	2.5	0.14	winter	0.16	1.2	2.7	0.21	0.59	1.75
	spring batch	0.32	1.1	0.13	0.43	4.3	0.16	spring	0.13	0.53	0.89	0.29	0.69	0.55
	summer batch	1.0	2.2	0.48	1.1	1.9	0.16	summer	0.18	0.22	0.14	0.13	0.23	0.42
	fall batch	0.30	5.7	0.33	0.29	13.0	0.49	fall	0.22	0.14	0.21	0.20	0.04	0.14

*: big batch include all winter deployment for the entire transect plus all seasons for the northern two sites: Alert, Kuujjuaq and Fraserdale.

Table S11: Percentage of determined values that fall above the MDL and MQL.

	Analyzed by Dow			Analyzed by NILU		
	D4	D5	D6	D4	D5	D6
above MQL	82%	78%	37%	90%	75%	38%
between MDL and MQL	13%	15%	23%	8%	18%	32%
below MDL	5%	7%	40%	2%	7%	30%

In Table S8 and S9, different fonts are used to indicate whether a reported sorbent concentration is above the MDL and MQL. Levels above the MQL are presented in bold font, whereas levels between MDL and MQL are in italic font. Levels below the MDL are given in brackets. If the blank correction yielded a negative value, i.e., if the field blank contained more chemical than an exposed sampler, the tables contain an "ND" entry.

Please note that while the MDL and MQL are based on the standard deviation of the field blanks belonging to a particular batch, the blank correction was not done using the average of these field blanks. Instead, we thought it preferable to use the individual field blank matched with each exposed sampler for blank correction.

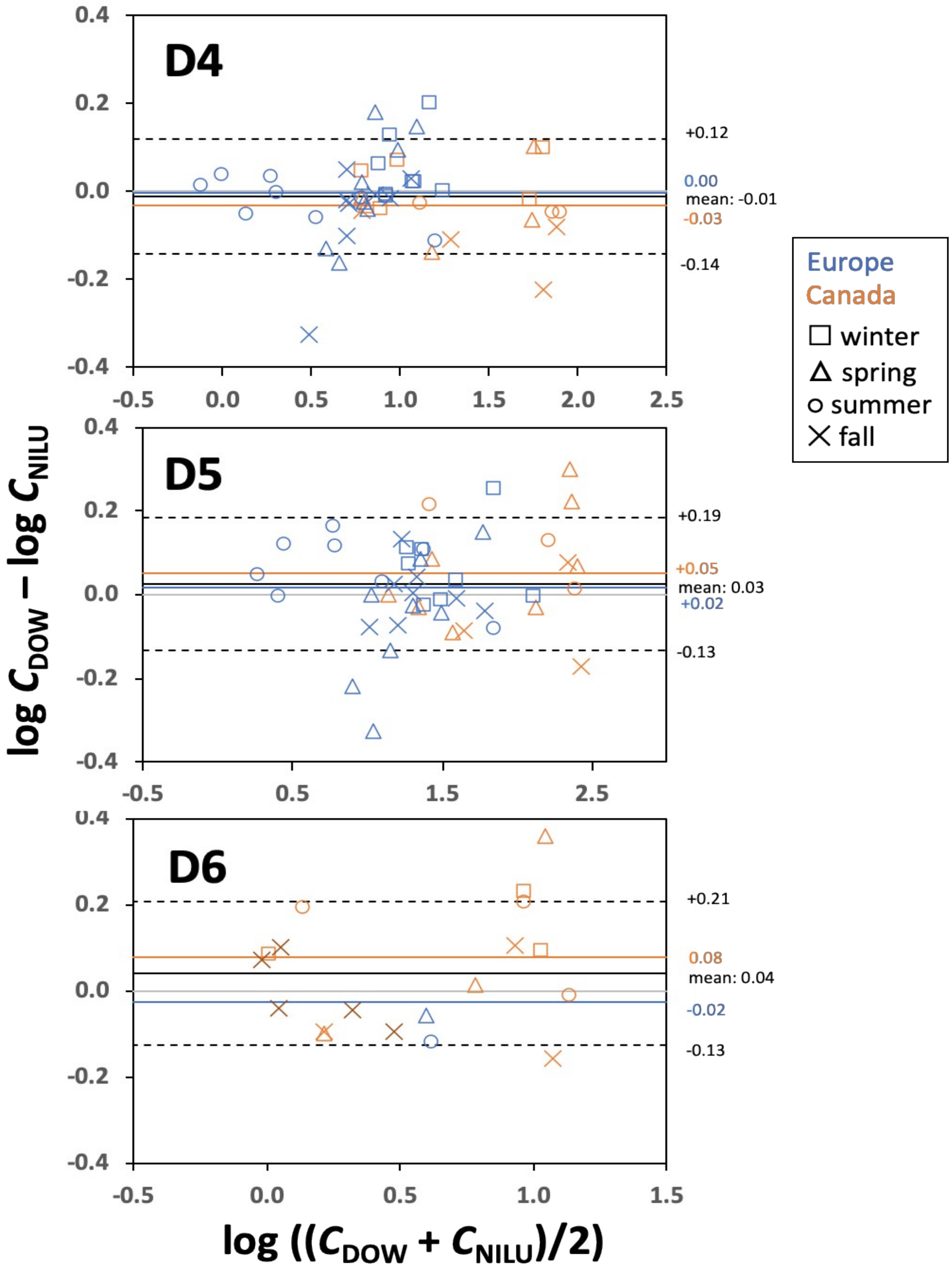

Figure S2: Bland Altman plot displaying the discrepancy between the blank-corrected levels in ng/g sorbent determined by Dow and NILU against their average. Both discrepancy and average are shown on a logarithmic scale. The numbers on the right-hand side indicate the overall mean bias, the mean bias of samplers deployed along the Canadian (orange) and European transect (blue) and the limits of agreement, which comprise the range in which the results from the two labs agree with each other 19 out of 20 times.

Table S12: Time averaged concentrations of cVMS in ng m^{-3} during four seasons at different sampling locations along the Canadian transect, measured by Dow and NILU. Bold font: > MQL, Italic font: > MDL, in brackets: < MDL, ND: blank correction gave negative value.

Season	Location	Dow			NILU		
		D4	D5	D6	D4	D5	D6
winter	Alert, Nunavut	*0.40*	*0.89*	(0.008)	**0.62**	*1.3*	*0.054*
	Kuujjuaq, Quebec	*0.74*	*1.8*	*(0.05)*	**1.1**	*2.1*	*0.059*
	Fraserdale, Ontario	**1.6**	**3.0**	*0.084*	**1.4**	*2.3*	*0.072*
	Algonquin Ontario	**1.8**	**5.2**	*0.17*	**2.0**	**5.7**	**0.15**
	Borden, Ontario	**2.5**	**7.2**	**0.27**	**2.2**	**5.9**	**0.23**
	Downsview, Ontario	**18**	**72**	**3.0**	**14**	**43**	**1.7**
	Toronto, Ontario	**13**	**67**	**3.0**	**14**	**57**	**2.4**
spring	Alert, Nunavut	*0.69*	*0.84*	(0.030)	**0.96**	*1.8*	*0.091*
	Kuujjuaq, Quebec	*0.62*	*0.91*	(0.022)	**0.94**	*1.8*	*0.052*
	Fraserdale, Ontario	**1.5**	*2.3*	(0.057)	**1.7**	**3.2**	*0.094*
	Algonquin Ontario	**1.5**	**3.5**	*0.11*	**1.5**	**3.5**	*0.15*
	Borden, Ontario	**3.2**	**8.1**	**0.37**	**4.4**	**9.9**	**0.44**
	Downsview, Ontario	**13**	**32**	**1.5**	**15**	**34**	**1.5**
	Toronto, Ontario	**16**	**74**	**3.7**	**12**	**37**	**1.7**
summer	Alert, Nunavut	(0.18)	(0.25)	(0.020)	(0.18)	(0.25)	(0.032)
	Kuujjuaq, Quebec	ND	(0.13)	(0.008)	*0.49*	(0.24)	(0.027)
	Fraserdale, Ontario	*0.52*	*1.1*	(0.030)	*0.57*	*1.0*	*0.049*
	Algonquin Ontario	*0.89*	*1.9*	(0.037)	*0.86*	*1.2*	*0.069*
	Borden, Ontario	**3.2**	**8.1**	**0.42**	**3.2**	**4.9**	**0.27**
	Downsview, Ontario	**17**	**46**	**2.7**	**19**	**34**	**1.7**
	Toronto, Ontario	**19**	**62**	**3.5**	**21**	**60**	**3.5**
fall	Alert, Nunavut	*0.42*	(0.77)	(0.0067)	*0.57*	*1.7*	*0.052*
	Kuujjuaq, Quebec	(0.072)	ND	ND	*0.20*	(0.23)	(0.011)
	Fraserdale, Ontario	*0.79*	*2.0*	(0.052)	**0.99**	*1.9*	*0.059*
	Algonquin Ontario	**1.4**	*4.0*	(0.057)	**1.6**	(3.2)	(0.077)
	Borden, Ontario	**4.2**	**9.9**	**0.37**	**5.4**	**12**	**0.44**
	Downsview, Ontario	**17**	**59**	**2.4**	**21**	**49**	**1.9**
	Toronto, Ontario	**12**	**53**	**2.4**	**20**	**80**	**3.5**

Table S13: Time averaged concentrations of cVMS in ng m^{-3} during four seasons at different sampling locations along the European transect, measured by Dow and NILU. Bold font: > MQL, Italic font: > MDL, in brackets: < MDL, ND: blank correction gave negative value.

Season	Location	Dow			NILU		
		D4	D5	D6	D4	D5	D6
winter	Zeppelin, Svalbard*	**2.5**	**6.4**	(0.30)	**1.9**	**4.9**	(0.022)
	Gortinak, Norway	**3.0**	**5.2**	(0.067)	**2.7**	**4.0**	(0.15)
	Andøya, Norway	**3.0**	**10**	*0.91*	**3.0**	**9.4**	(0.22)
	Tustervatn, Norway	**2.0**	**5.2**	(0.22)	**1.7**	**4.2**	(0.20)
	Kårvatn, Norway	**2.0**	**5.7**	(0.42)	**2.1**	**6.2**	(0.54)
	Birkenes, Norway	**2.1**	**7.7**	(0.16)	**2.1**	**7.9**	(0.47)
	Blæsbjerg, Denmark	**4.4**	**22**	*1.1*	**2.7**	**12**	(0.40)
	Weilerswist, Germany	**4.2**	**32**	*1.4*	**4.2**	**32**	*1.3*
spring	Zeppelin, Svalbard	**1.5**	**2.7**	(0.10)	**1.6**	**2.7**	(0.040)
	Gortinak, Norway	**1.5**	**3.0**	(0.091)	**1.7**	**4.0**	(0.12)
	Andøya, Norway	**2.7**	**6.2**	*0.32*	**2.1**	**4.9**	*0.17*
	Tustervatn, Norway	**0.81**	**1.5**	ND	**1.1**	**2.5**	*0.17*
	Kårvatn, Norway	**0.91**	**1.7**	(0.10)	**1.3**	**3.7**	*0.23*
	Birkenes, Norway	**1.6**	**4.9**	(0.25)	**1.5**	**5.2**	*0.32*
	Blæsbjerg, Denmark	**2.1**	**7.2**	*0.35*	**1.4**	**7.9**	**0.59**
	Weilerswist, Germany	**3.5**	**17**	**0.91**	**2.5**	**12**	**1.0**
summer	Zeppelin, Svalbard	**0.32**	**0.49**	(0.013)	**0.35**	**0.44**	(0.020)
	Gortinak, Norway	**0.19**	**0.62**	*0.084*	**0.17**	**0.62**	ND
	Andøya, Norway	**0.49**	**1.8**	*0.10*	**0.44**	**1.2**	(0.079)
	Tustervatn, Norway	**0.25**	**0.79**	**0.19**	**0.22**	**0.59**	(0.008)
	Kårvatn, Norway	**0.49**	**1.7**	**0.16**	**0.49**	**1.3**	(0.11)
	Birkenes, Norway	**0.79**	**3.2**	**0.20**	**0.89**	**3.0**	*0.23*
	Blæsbjerg, Denmark	**1.5**	**6.7**	**0.42**	**1.5**	**5.2**	*0.35*
	Weilerswist, Germany	**3.5**	**16**	**0.89**	**4.4**	**19**	**1.2**
fall	Zeppelin, Svalbard	**1.2**	**4.0**	*0.19*	**1.1**	**3.7**	**0.17**
	Gortinak, Norway	**0.49**	**2.2**	*0.086*	**1.0**	**2.7**	**0.15**
	Andøya, Norway	**1.8**	**5.4**	**0.25**	**1.8**	**4.9**	**0.21**
	Tustervatn, Norway	**1.1**	**3.7**	*0.17*	**1.4**	**4.2**	**0.22**
	Kårvatn, Norway	**1.3**	**4.7**	**0.32**	**1.2**	**3.5**	**0.24**
	Birkenes, Norway	**1.2**	**4.9**	**0.27**	**1.3**	**4.9**	**0.30**
	Blæsbjerg, Denmark	**2.1**	**9.6**	**0.49**	**2.2**	**9.9**	**0.54**
	Weilerswist, Germany	**3.0**	**15**	**0.67**	**2.7**	**16**	**0.81**

*: Winter sampler deployment was not successful at Zeppelin in 2018. The data shown here were from a deployment between January 16 to April 1, 2020

Text S5: Comparison with Earlier Measurements

Although no comparable monitoring data exist for all sites along the sampling transects examined in this study, earlier studies have reported air concentration data for three of the sampling locations (a site representative of source regions and the two Northern terminals of the transects). Krogseth et al.[17] and Ahrens et al.[21] have measured cVMS in Toronto, Genualdi et al.[24] have reported a value for Alert, and Krogseth et al.[3], Bohlin-Nizzetto et al.[37-39], Warner et al.[31] have reported cVMS concentrations for the Zeppelin site, and Rauert et al.[40] have reported cVMS concentrations for Downsview, Alert and Ny-Ålesund (close to the Zeppelin site). Those data are compiled in Tables S14.

Table S14a: Comparison of the cVMS concentrations (ng m^{-3}) in Toronto measured here with literature values.

Time	D4	D5	D6	Comments	Source
Yearly	17±2 16±3	46±12 61±5	2.0±0.4 2.9±0.2	Downsview, XAD-PAS Toronto, XAD-PAS	This study
	21.4±2.5	142.6±6.7	11.5±1.0	Downsview, HiVol-AAS	21
Spring	14 14	33 55	1.5 2.7	Downsview, XAD-PAS Toronto, XAD-PAS	This study
	24.2±19.1	93.5±65.9	5.5±3.8	Scarborough, SPE-AAS	17
	14.6±2.3	85.6±21.4	2.9±0.2	Downsview, HiVol-AAS	21
	45±14	141±45	12±3	Downsview, SIP-PAS	40
Summer	18 20	40 61	2.3 3.4	Downsview, XAD-PAS Toronto, XAD-PAS	This study
	41±12	122±39		21 urban sites, XAD-PAS, July to October 2012	17
	11.2±1.6	67.5±7.6	7.7±0.7	Downsview, HiVol-AAS	21
	41	113	8.8	Downsview, SIP-PAS	40
Winter	16 13	58 62	2.3 2.6	Downsview, XAD-PAS Toronto, XAD-PAS	This study
	13.9±3.5	85.6±21.4	5.6±1.4	Downsview, HiVol-AAS	21
	22	85	5.9	Downsview, SIP-PAS	40

Ahrens et al.[21] measured at the Downsview site of the current study using a high-volume active air sampler, whereas Krogseth et al.[17] used the same sampling techniques employed within this study to measure summertime concentrations at 21 sites across Toronto. These two studies were conducted in 2012 and 2010/1, respectively, i.e., about a decade earlier than the current study. In general, levels measured in Toronto back then were approximately two (D4 and D5) or three times (D6) higher compared to those measured in the current study. The exception is the D4 concentration reported by Ahrens et al.[21], which is similar to the D4 concentration measured here. Interestingly, the D5/D4 concentration ratio in the data reported by Ahrens et al.[21] is more than double that measured within this study, whereas D5/D4 ratio reported by Krogseth et al.[17] was similar to that in the current study. This could suggest breakthrough loss of D4 in the HiVol technique employed by Ahrens et al.[21]. Overall, the data indicate that air concentrations in Toronto over the last decade declined by a factor of approximately 2 to 2.5, probably as a result of the reduced emissions. The measurements by Rauert et al., who sampled in 2013 and 2015, were found to be consistently higher compared to the current and previous studies

For Alert, there are two previous studies using a sorbent impregnated polyurethane foam PAS, one with only one measurement.[24,40] The mean concentrations in Rauert et al. were at least one order of magnitude greater than in this study with variability being very high.[40] This could be a consequence of sample contamination. The average D5 concentrations of 0.78 ng/m^3 for spring and summer within this study was similar to the value of 0.58 ng/m^3 reported by Genualdi et al.[24] However, the D6 concentration reported by Genualdi et al.[24] of 0.31 ng/m^3 is much higher than the value of 0.04 ng/m^3 reported here. The D5/D6 ratio of this earlier measurement appears as an extreme outlier compared to other atmospheric data reported in the literature. This suggests that the D6 value in Genualdi et al. may also be compromised by sample contamination.

Table S14b: Comparison of the cVMS concentrations (ng m^{-3}) in Alert measured here with literature values.

	Time	**D4**	**D5**	**D6**	**Comments**	**Source**
Alert	Yearly	0.51±0.26	0.97±0.49	0.04±0.02	XAD-PAS	This study
	Spring-Summer	0.51	0.78	0.04	XAD-PAS	This study
			0.58	0.31	SIP-PAS	24
		21±20	10±14	13±20	SIP-PAS	40

For Zeppelin in the European Arctic, our measured values of D4 and D5 were slightly higher than values reported for earlier years, but not statistically different. The D6 concentration at this site was lower than values for previous years (2016 and before) reported by others, particularly during Spring-Summer. The data from Rauert et al. were once again much higher than reported in the other studies. Higher measurements reported on SIP-PAS could be attributed cVMS stability on these samplers over long deployment times. However, the same sorbent used on SIP-PAS (i.e., XAD-2) was also used within the current study, where no degradation was observed to occur over the time range of sample deployment. This would suggest contamination either through sample work up or from the PUF material itself is contributing to the consistent higher observations reported by Rauert et al.

Table S14c: Comparison of the cVMS concentrations (ng m^{-3}) in Zeppelin station measured here with literature values.

	Time	D4	D5	D6	Comments	Source
Zeppelin	Yearly	1.3±0.8	3.1±2.2	0.13±0.11	XAD-PAS	This study
	Fall-Winter	1.7±0.7	4.7±1.4	0.22±0.06	XAD-PAS	This study
		0.68±0.10	2.73±0.24	0.42±0.06		3
		1.49±0.24	3.60±0.67	0.39±0.08		37
		1.14±0.12	3.66±0.48	0.39±0.09		38
		1.48±0.04	2.93±0.19	0.15±0.02		30
		67	25	3.8	Ny-Ålesund, SIP-PAS	40
	Spring-Summer	0.94±0.86	1.54±1.53	0.04±0.04	XAD-PAS	This study
		0.26±0.02	0.75±0.09	0.24±0.05		3
		0.48±0.07	0.72±0.10	0.87±0.10		37
		0.93	1.50	1.91		38
		0.15±0.02	0.14±0.03	0.07±0.01		30
		25±10	6.5±0.14	1.45±0.21	Ny-Ålesund, SIP-PAS	40

In summary, the comparison with air concentrations previously reported for Toronto suggests that levels have declined by a factor of approximately 2 to 2.5 over the last decade. Levels of D4 and D5 measured at the Arctic endpoints of the two transects are similar to those reported previously for Zeppelin and Alert, but D6 levels tended to be lower than values reported earlier.

New references

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40. C. Rauert, J. K. Schuster, A. Eng, T., Harner. 2018. Global atmospheric concentrations of brominated and chlorinated flame retardants and organophosphate esters. Environ. Sci. Technol., 52(5), pp.2777-2789.

Text S6: Comparison between Field Blanks measured at NILU and Dow

While Figure 2 illustrates that field blank levels determined by the two labs are generally in good agreement, we further explored this agreement quantitatively by plotting the discrepancy in the field blank levels from Dow and NILU against their average (i.e., in a type of Bland Altman plot) (Figure S3). As environmental concentration data are typically log-normally distributed, both the discrepancy and the average are displayed on a log scale. We note that there is no a priori reason why the field blanks in the two labs should be identical. The samplers underwent different transport routes to reach the two labs and also the processing and extraction of the samples in the labs may lead to differences in field blank contamination.

The Figures S3 and S4 illustrate again the large difference in the D5 and D6 blank levels of the European and Canadian transect samples (D5 lower in EU, D6 lower in NA). It also shows the unusually high D6 field blank contamination in the winter deployments in the Canadian transect. In general, there is slight positive bias, indicating that the field blank levels were generally higher during Dow's analysis than NILU's. This bias increases from 0.03 log units for D4 to 0.08 log units for D5 to 0.14 log units for D6. However, there is no general tendency for the bias to be consistently higher for North American or European samples. A bias of 0.14 log units means that Dow's blank levels for D6 are on average 1.4 times greater than those reported by NILU, which is still a small difference considering the variation observed during the quantification of small quantities close to the limits of detection. The plots in Figure S3 provide no indication that the discrepancy between field blank level or the bias is dependent on the field blank concentrations. The limits of agreement are ±0.32 log units for D4 and D5 and ±0.54 log units for D6, i.e., there is a 95 % likelihood that the field blank levels determined by the two labs are within a factor of 2.1 or 3.5 of each other.

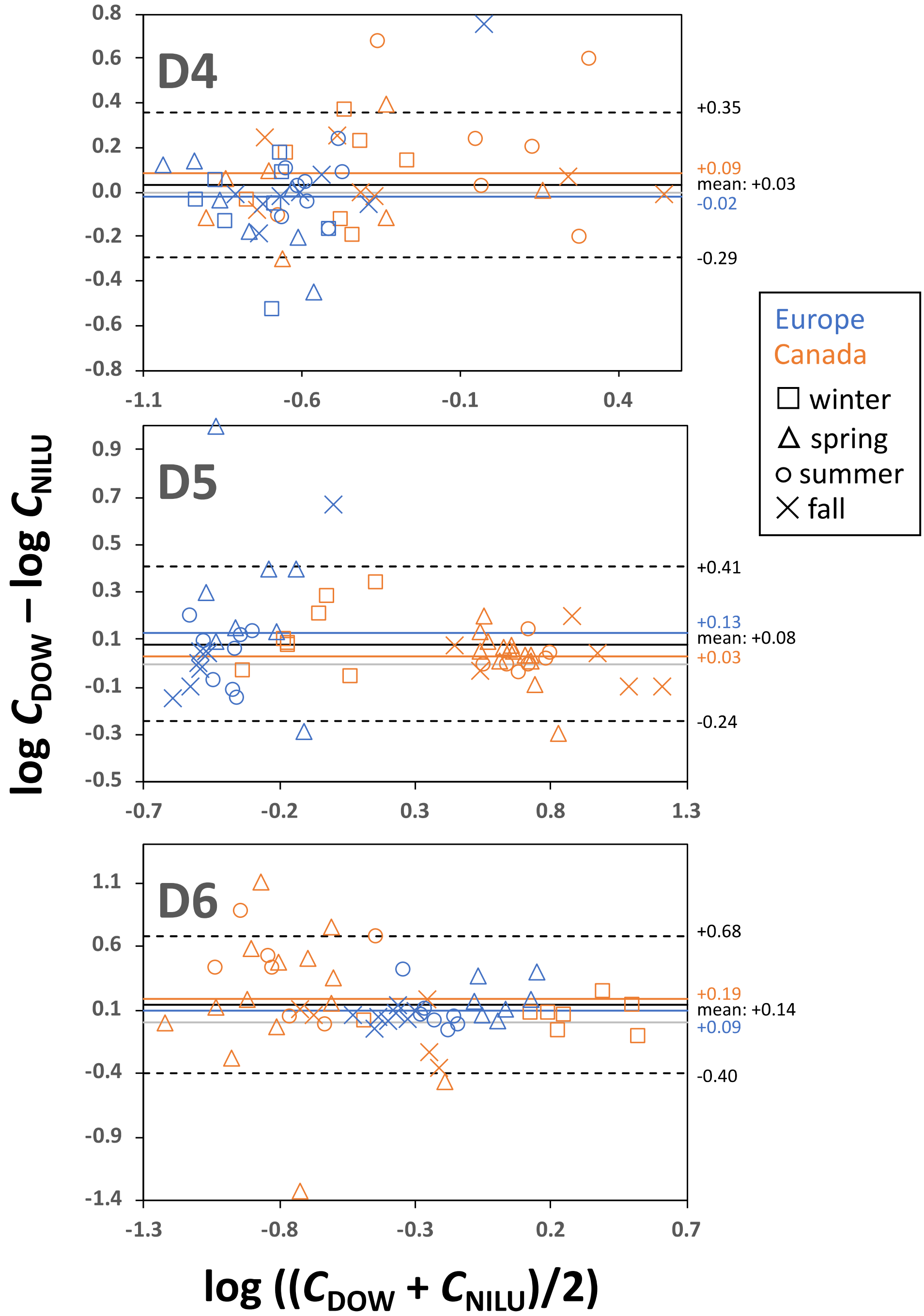


Figure S3: Bland Altman plot displaying the discrepancy between the field blank levels in ng/g sorbent determined by Dow and NILU against their average. Both discrepancy and average are shown on a logarithmic scale. The numbers on the right-hand side indicate the overall mean bias, the mean bias of samplers deployed along the Canadian (orange) and European transect (blue) and the limits of agreement, which comprise the range in which the results from the two labs agree with each other 19 out of 20 times.

Figure S4: Field blanks for different sites along the Canadian transect based on the batches of resins: All open columns were from the first big batch; Solid blue, red and light brown bars representing the second, third and fourth batches. Data from left two panels were from Dow and right two panels from NILU.

APPENDIX: Instruction on Sampler Deployment and Retrieval Send to Local Contact People

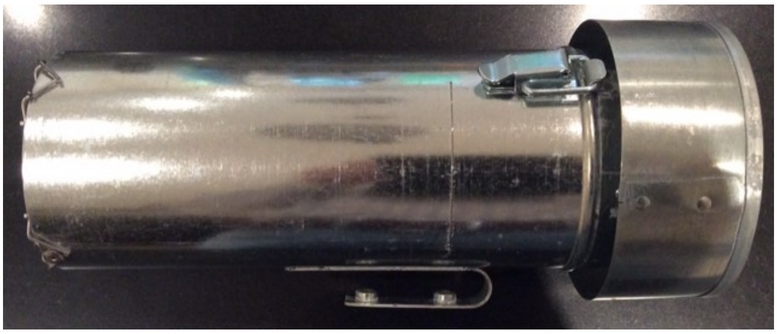
Introduction

Thank you for agreeing to help with our project which is using passive air samplers to gain insight in the ability of different chemicals to undergo long range atmospheric transport. **Your sampling site is one of 16**, half of which are in North America and the other half in Europe. On both continents, the eight sampling sites extend over a wide range of latitudes from Southern Canada to Ellesmere Island and from Central Europe to Svalbard. **Deployment periods at the different sites should be synchronized as much as possible**, i.e. dates of deployment and retrieval should be more or less the same at all 16 sites.

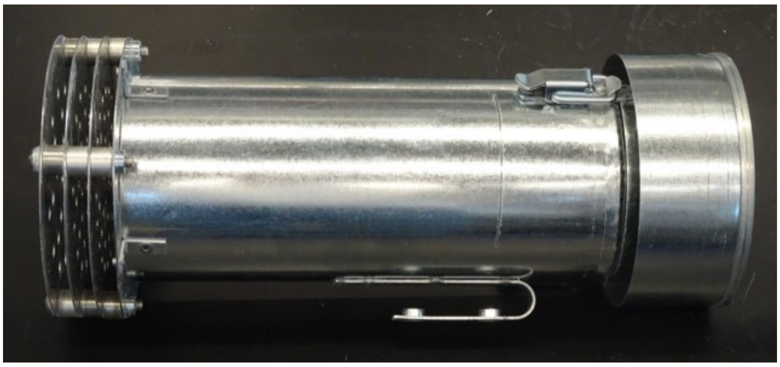
We plan to sample a group of compounds called volatile methyl-siloxanes (VMS) for **four consecutive three-month periods for a total sampling time of one year**.

The sampling material comprises:

- A **stainless-steel sampler housing**, which consists of a bottom part and a cap, which is attached to the bottom with the help of toggle latches. The brace attached to the side of the bottom part can be used to attach the housing to a solid support structure.

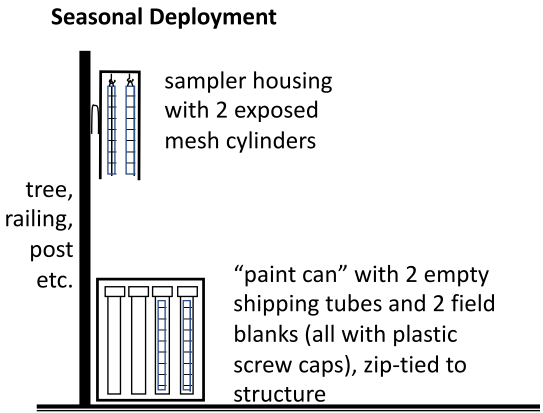


Northern sites, which may experience high winds, will be supplied with a housing that has a modified bottom to prevent wind blowing into the housing.



- For each of the four seasonal deployment periods you will be supplied with a metal “paint can” (left) containing **4 cylindrical, resin-filled stainless-steel mesh cylinders** (right bottom), which are individually stored in **metal shipping tubes closed with a plastic cap** (right top). The paint can also contains nitrile gloves, a number of aluminum foil squares and Teflon tape (only in the can for the first seasonal deployment) for sealing the shipping tubes.


- **Each seasonal deployment is duplicated and has two associated field blanks**. Because duplicates are placed in the same sampler housing at the same time, only one housing is required.
- During a deployment period, the sampling site should look like this:



We aim to start the first deployment in mid-November 2018. The sampling schedule therefore looks like this:

2018		2019										
November	December	January	February	March	April	May	June	July	August	September	October	November
period 1 - duplicate 1			period 2 - duplicate 1			period 3 - duplicate 1			period 4 - duplicate 1			
period 1 - duplicate 2			period 2 - duplicate 2			period 3 - duplicate 2			period 4 - duplicate 2			
period 1 - field blank 1			period 2 - field blank 1			period 3 - field blank 1			period 4 - field blank 1			
period 1 - field blank 2			period 2 - field blank 2			period 3 - field blank 2			period 4 - field blank 2			

If the installation of the sampler housings and the deployment of the first set of mesh cylinders is done during the same visit, you should be able to perform all required tasks during **five visits to the sampling site**, in mid-November 2018, mid-February 2019, mid-May 2019, mid-August 2019 and mid-November 2019. I will contact you by e-mail two weeks prior to each sample changeover, as well as a few days before that date.

The VMSs, i.e. the group of compounds that we are studying, are present in most personal care products (e.g. shampoo, deodorant, skin cream, etc.). It is imperative that we eliminate the possibility of sample contamination during deployment and retrieval procedures. This requires that anyone visiting the sampling site do not use such products on the day of the visit. This means you cannot use any skin care products, take a shower or wash your hair prior to visiting the site. Also, do not any use or handle any other silicone containing materials (lubricants, sealants, etc.) on a day of sampling.

On the following pages are detailed instructions for:

- Installing the sampler housing (pages 4 to 5) and
- Deploying and retrieving samplers for seasonal deployments (pages 6 to 9)

Your contact for any questions or information is: Frank Wania, +1-416-516-6542 (h) or +1-416-287-7225 (w), frank.wania@utoronto.ca

Installation of the sampler housing

1. Selecting a suitable sampling site

The ideal sampling site:

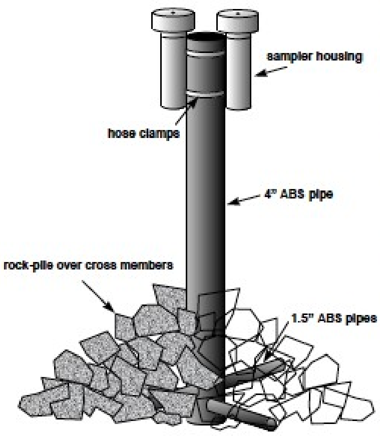
- is far from human activity
- can be safely visited during all times of the year
- is secure in the sense that it is unlikely anyone unauthorized will tamper with, or remove the sampler
- either has an existing structure to which the sampler housing can be attached, or it is feasible to erect such a structure

2. Structure to which sampler housing is attached

The sampler housing should be attached to a solid support structure, which could be a sturdy tree, a fence post, or a railing (examples are shown below). If there is no suitable existing structure, a temporary post could be installed. One option involves an L-shape metal rod, that is rammed into the ground and secured with aircraft cable wires and tent pegs (below right).




In latitudes with permafrost, structures relying on ABS pipes and stone cairns may be a better option (see below).



3. Attaching the housing to the structure

Attach the sampler housing to the support structure using several hose clamps or strong cable ties. The housing should be attached firmly, so it will not be blown away, not rattle in the wind, and remain upright during the entire deployment period.

Orientation

- The **open side of the housing is at the bottom**, the removable cap is at the top. The housing should be installed as straight as possible.
- If winds at the site can be strong and there is a prevailing wind direction, it is preferable to have the sampler deployed in such a way that it is not directly facing the wind.

Deployment height

- Aim to have the **middle of the sampler housing at a height of approximately 1.3 m** (chest height) above the ground. This height assures that there is minimal influence of the ground on the sampler, yet also allows for convenient sampler exchange without the use of a ladder.
- There may be good reasons for deviating from this sampler height, e.g. if snow depth can be appreciable at the site (the sampler housing should not be buried in the snow) or if it is desirable to have the sampler out of reach of curious passers-by (human or otherwise).

4. Record keeping

- Take a few photographs of the installed sampler housing, the sampling site and its immediate surroundings.
- Note the geographical coordinates of the sampling location (e.g. using a GPS or compass in your phone or car/snowmobile), so that we have a record of it and you can find it again.

Checklist before heading out to install the sampler housing

Do not use personal care products (shampoo, deodorant, skin cream, etc.) on the day of your visit to the sampling site.

Make sure you have with you:

- a set of silicone-free laboratory gloves
- the sampler housing
- a number of hose clamps, copper wires or cable ties
- the tool (e.g. screwdriver) you need to tighten the hose clamps
- a pair of scissors to cut the end of the cable ties

If you install a post, you may also need:

- a post
- an appropriate length of aircraft cable wire and aluminum sleeves of the correct size
- the tool(s) required for (i) squeezing those sleeves and (ii) cut the wire
- tent pegs (3 per post)

Deployment and Retrieval of Samplers for Seasonal Deployments

There are up to three procedures to perform during a sampling site visit:

A. Retrieval of Exposed Mesh Cylinders
Expose the two "old" field blanks briefly to the atmosphere. Take currently exposed mesh cylinders out of the sampler housing and place them into shipping tubes. Send those four tubes for analysis.

B. Deployment of New Field Blank
Expose the two "new" field blanks briefly to the atmosphere and store on-site.

C. Deployment of New Mesh Cylinders
Place two mesh cylinders into sampler housing and store the corresponding, empty shipping tubes on-site.

The sequence in the procedures is always A before B before C. **During the first visit, only procedure B and C are required.** During the second, third and fourth visit, all three procedures are required. During the fifth and final visit, only procedure A is required.

Checklist before heading out to deploy, exchange or retrieve samplers

Do not use personal care products (shampoo, deodorant, skin cream, etc.) on the day of your visit to the sampling site.

Make sure you have with you:

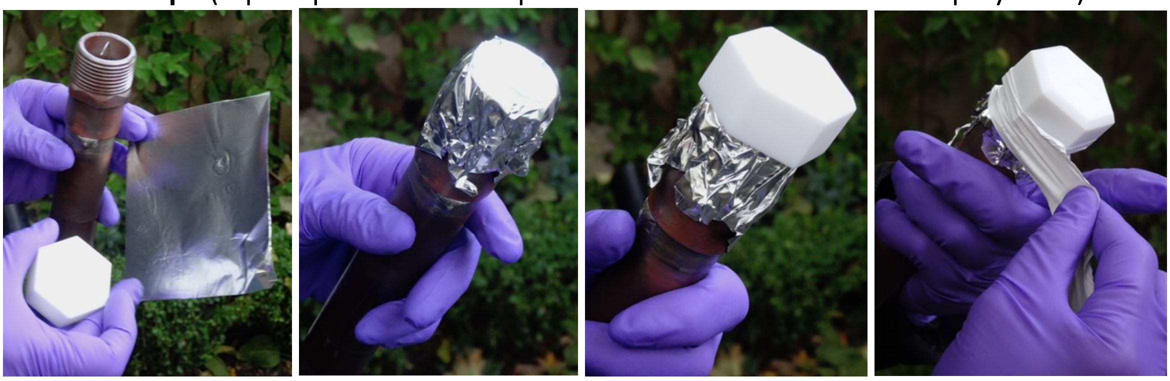
- The paint can for the appropriate seasonal deployment, containing:
  - A set of silicone-free laboratory gloves
  - Four shipping tubes with clean mesh cylinders
  - Teflon tape for sealing the shipping containers (only provided in the can for the first deployment)
  - Aluminum foil squares
- The paint can opener (supplied in the box for the first seasonal deployment) and a tool for closing the paint can (e.g. a hammer)
- Water-proof felt pen for labelling samplers
- Zip-ties or copper wire for securing field blanks and empty shipping containers on-site
- Pair of scissors to cut zip-ties

1. Open the paint can (using the tool supplied in the box for the first seasonal deployment) and **wear the laboratory gloves that are in the can.**

A. Retrieval of Exposed Mesh Cylinders (this step NOT required during 1st deployment visit)

2. Retrieve (from where you had stored them) the two empty shipping tubes corresponding to the mesh cylinders currently deployed in the sampler housing and open them.
3. Open the toggle latches that attach the cap of the housing to the bottom of the sampler housing.
4. Lift the cap from the housing, with the mesh cylinders dangling from the hooks inside the cap (see below left).

5. Place the dangling mesh cylinders into the shipping tubes (see above right) and unhook them from the cap. If the mesh cylinders got unhooked during the deployment period, they will rest in the bottom of the housing. Use you **gloved** hands to move them from the housing into the shipping tube. Make a note “unhooked” on the label if this was the case.
6. With the retrieved mesh cylinders inside, close the two **shipping tubes by (i) placing one of the provided aluminum foil squares over the opening, (ii) screwing the white Teflon plastic cap tightly, and (iii) sealing the gap between cap and tube by wrapping it with Teflon tape** (tape is provided in the paint can for the first seasonal deployment).



7. Label each of the retrieved samplers. Specifically indicate the date of retrieval.
8. Retrieve the field blanks that had been placed at the sampling site for the past deployment period, open them and take the mesh tubes out (using the hooks in the housing cap, see picture below). After 1 minute, place them back into the shipping tubes, and cap them

tightly (following the instructions of step 6 above). Add the retrieval date to the label. Always keep and ship them together with the two retrieved samplers.

9. Place the four tubes into a paint can, close it, place in a cardboard box and ship as soon as possible to: Frank Wania, University of Toronto Scarborough, Department of Physical and Environmental Sciences, 1265 Military Trail, Toronto, Ontario, Canada M1C 1A4, Tel. 416-287-7225, frank.wania@utoronto.ca.

   With the shipment include a description of anything that may have been out-of-the-ordinary. Also include the name of the person performing the deployment and retrieval.

B. Deployment of New Field Blanks

10. Take two of the shipping tubes with clean mesh cylinders out of the paint can for the next seasonal deployment and remove the caps.

11. Slide the two mesh cylinders out of the shipping tubes by attaching a hook on the inside of the housing cap to the metal wire at the top of the mesh cylinder (see picture below left). Leave exposed to the air for 1 minute (below right).

12. After 1 minute, place them back into the same shipping tubes, **cap the shipping tubes tightly** (following the instructions of step 6 above).

13. Label the shipping tube with “field blank” and the current date. Place the two shipping tubes into the paint can.

C. Deployment of New Mesh Cylinders

14. Take the remaining two shipping tubes with clean mesh cylinders out of the paint can for the next seasonal deployment and remove the caps.
15. Attach two of the hooks on the inside of the housing cap to the metal wires at the top of the mesh cylinders and slide the mesh cylinders out of the shipping tubes.
16. Place the mesh cylinders into the sampler housing and close the sampler housing using the toggle latches (see below). There are guide wires inside the housing to limit the movement of the mesh cylinders within the housing. Careful that the mesh cylinder do not get unhooked during this process. If they do, use your gloved hands to reattach them to the hook.

17. Close the empty shipping tubes and label them with the current date.
18. Add these two shipping tubes to the two field blanks in the paint can (see step 13), zip-tie to a structure close to the sampling site in the open.