Supporting Information for

Temporal Trends of Halogenated Flame Retardants in the Atmosphere of the Canadian Great Lakes Basin (2005-2014)

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Table of Content **Page**

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Section S1. Sample Collection, Analysis Methods and Data Management

S1.1 Air Sample Collection

Air was pulled through a General Metal Works PS-1 high volume sampler at ~0.24 m^3 min^{-1}, for a total volume of about 350 m^3 over a 24-hour sampling period. Particles were collected on a prebaked (400 °C) 10.2 cm diameter glass fiber filter (GFF, Gelman A/E Microfiber) and vapor phase organic pollutants were collected on a pre-cleaned polyurethane foam (PUF) (7.5 cm length x 6.2 cm diameter; PUF, Pacwill Environmental) plug.

S1.2 Air Sample Analysis

Air samples were analyzed in the Organics Analysis Laboratory (OAL), Environment and Climate Change Canada, Toronto, ON, Canada. The PUF plugs were Soxhlet extracted using 475 mL hexane for 24 hour. The PUF extracts were dried using pre-cleaned anhydrous sodium sulfate, reduced to 2 mL in iso-octane keeper using a Zymark/Biotage Turbovap 500 system at 38 °C. The PUF extracts were divided into 2 aliquots of 1 mL each. The GFFs were extracted with a Dionex ASE200 using a hexane/acetone mixture (7:3), 2000 psi, 100 °C, 5 minute static cycle, 3 cycles per sample, 150% flush volume, 60 second purge with UHP nitrogen. The GFF extracts were reduced to 1 mL in iso-octane keeper using a Biotage Turbovap 200 system at 38 ^{o}C under a nitrogen pressure setting of 7 psi. The GFF extracts were not dried prior to volume reduction. For PBDE and non-BDE FR analysis, one PUF aliquot and the GFF extract were cleaned separately using silica columns on a Microlab 4000 automated SPE system with 6 mL of dichloromethane to remove interferences. All the SPE extracts were concentrated on the Zymark/Biotage Turbovap 200 system at 38 ^{o}C, a nitrogen pressure setting of 6 psi, with acetonitrile keeper to 0.5mL then accurately adjusted to 1 mL.

A 15 m x 0.25 mm i.d. x 0.10 μm film thickness Restek Rtx-1614 column was used with the GC-ECD (Agilent 6890) for determination of BDE-209 and DBDPE. The oven temperature used was 100 ^{0}C for 1 min, 25^{0}C/min to 200^{0}C then 4.25^{0}C/min to 315^{0}C and hold for 3 min. Ultra high purity (UHP) He (Praxair) was used as carrier gas in constant-flow mode at a column flow-rate of approximately 1.2 mL/min. Pulsed splitless-mode injections of 1 μL with pulse pressure of 30 psi for 1.05 min (purge on at 1.0 min) were made using an autosampler/autoinjector system (Agilent 7683B series). Injection port and detector were held at 250^{0}C and 350^{0}C respectively. ECD make-up gas was UHP N_2 (Praxair) set at constant flow of 30 mL/min. Data files were manually integrated and processed using external standard methods for identification and quantification.

Gas chromatography electron capture negative ion mass spectrometry (GC-ECNIMS) was used to provide confirmatory analysis of processed sample fractions for all target analytes, except for BDE-209 and DBDPE. An Agilent MS-Engine system was used for samples up to and including the 2008 sampling year. An Agilent 5975 MS system was used for samples from 2009 onwards. 2009 samples were analysed using both the Agilent MS-Engine and the Agilent 5975 MS. Data from both methods were compared and good agreement was found. The coefficient of variance between the two methods ranged from 0 to 25 %.

The MS-Engine system instrumentation consisted of an HP5890 GC interfaced to an HP5989B MS (MS-Engine) mass spectrometer, equipped with negative ion chemical ionization capability and high-energy

dynode (HED) detector. Gas chromatographic conditions consisted of a heated splitless/split injection port (Merlin micro-seal; deactivated, borosilicate glass, single tapered liner) at a temperature of 200^{0}C; a 30 m x 0.25 mm i.d. x 0.25 μm film thickness DB5 column (J&W Scientific) was used (80^{0}C for 2 min, 15^{0}C/min to 160^{0}C then 2.5^{0}C/min to 260^{0}C and hold for 62 minutes), trace analytical grade (TAG) He (Praxair) as carrier gas in constant-pressure mode, at an initial column flow-rate of approximately 1.3 mL/min. Splitless-mode injections of 2 μL (purge on at 2.0 min) were made using an autosampler/autoinjector system (HP 7673 series). Mass spectrometer operational conditions were: transfer line, 275^{0}C; source, 150^{0}C; quadrupole, 100^{0}C; reagent gas, UHP methane (Praxair) at 2.0 torr; electron energy, 230 eV; emission current, 300 mA; electron multiplier voltage (typical tune plus 500V), HED voltage, 10,000 V. Selected ion mode (SIM) was employed to maximize sensitivity for PBDEs, with just two ions being monitored: m/e at 79 for quantitation (Q) and m/e at 81 for confirmation (C) with dwell time set at 100 ms. At appropriate points in the chromatogram, representative ions for the retention markers TBB (314) and OCN (404) were monitored. Overall system control, data acquisition and data reduction was conducted with an HP59944C MS ChemSystem (UNIX). Data files were manually integrated and processed using external standard methods for identification and quantification.

The Agilent 5975 MS system instrumentation consisted of an Agilent 7890A GC interfaced to an Agilent 5975 MSD equipped with negative ion capability. GC conditions consisted of a heated splitless/split injection port at a temperature of 200ºC; a 30 m × 0.25 mm i.d. × 0.25 μm film thickness DB5 column (J&W Scientific) was used (80ºC for 2 min, 15ºC/min to 160ºC then 2.5ºC/min to 260ºC for 4 min then 2.5ºC/min to 285ºC and hold for 15 min). Pulsed splitless-mode injections of 1.5 μL were made using an autosampler/autoinjector system (Agilent 7683 series). An injector pulse pressure of 35 psi was used until 1.5 mins. A purge flow to split vent of 50 mL/min at 1.0 min was utilized. MS operational conditions were: transfer line, 280ºC; source, 150ºC; quadrupole, 150ºC; reagent gas, UHP methane (Praxair) a flow rate of 40 % (the value is a percentage of maximum flow rate). Selected ion mode (SIM) was employed to maximize sensitivity for PBDEs and BFRs, with the following ions being monitored: m/e at 79, 81, 160, 237, 291, 404, 464, 551.5 and 652. Overall system control, data acquisition and data reduction was conducted with an Enhanced ChemStation and data files were manually integrated and processed using external standard methods for identification and quantification.

S1.3 Air Sample Blanks and Recoveries

Six field blanks were collected each year at every site by placing a sampler head with a PUF and a GFF in the HiVol sampler and immediately removing without drawing air through the sampler. Laboratory and field blanks were processed in the same way as the samples to determine the contamination introduced during extraction and clean-up; and by handling, shipping and storage, respectively. Laboratory blanks were included with every second batch of samples. A blank clean PUF and a blank clean GFF were spiked with 1.00 mL of halogenated flame retardant (HFR) standard (10 pg/μL for each HFR), and processed as air samples. For each sample batch, a silica column was spiked with 1.00 mL of the same HFR standard and taken through the SPE cleanup procedure.

S1.4 Data Management

Research Data Management and Quality Control System^{TM} (RDMQ^{TM}) is an interactive SAS-based system which provides a means of evaluating and flagging data (Sukloff et al., 1995). The RDMQ^{TM} evaluates raw

data by assigning flags based on sample quality (duration and volume of sample, meteorological conditions, presence of unusual anthropological or natural conditions at the site, and adherence to field and laboratory protocols). Data are flagged and may be invalidated if they are of low sample quality. Statistical analysis of field blank concentrations was used to assess for background method concentrations and to determine method detection limits. Statistical analysis of samples were used to assess the percentage of quantifiable samples and to identify and investigate outliers. The data were also manually reviewed and identified for any anomalies present by plotting time series of ambient concentrations as well as quality indicators such as blank levels and method detection limits. Finally, summaries of quality control activities such as the analysis of common reference standards and matrix spikes were used to provide information about the quality of the final dataset. The summarized data were approved by the data management team and the principal investigator. The raw data have undergone several evaluation steps before being accepted as valid data.

Section S2. Ratios between the dominant PBDE congeners (BDE-47 and -99) and *anti*- DDC-CO and total DDC-CO

BDE-47, -99 and -100 are the main components of the commercial Penta-BDE formulations, with BDE-47 accounting for 38-43 %, BDE-99 account for 45-49 % and BDE-100 accounting for 7.8-13% of the product[1]. Pearson correlation analysis was performed on the concentrations of the main components of Penta-BDE technical mixture to determine whether they were from the same source. Significant correlations were observed among BDE-47, -99 and -100 (Table S4), suggesting that they likely came from similar sources. The yearly average ratio of BDE-47/-99 in the GLB air samples ranged from 0.9 to 3.3 (Table S5), which are higher than the ratios measured in the commercial Penta-BDE mixtures (DE-71 = 0.79 and Bromkal 70-5DE = 0.96), whereas the yearly average ratios of BDE-99/-100 (ranged from 3.5 to 6.1) are similar to those ratios measured in the Penta-BDE formulations (DE-71 = 3.7 and Bromkal 70-5DE = 5.7). Su et al.[2] also reported similar BDE-47/-99 and BDE-99/-100 ratios in the air samples collected at Point Petre. Photo-oxidation during LRAT can modify the composition of these mixtures; the photo-degradation half-lives of BDE-47 and -100 are longer than that of BDE99[3]. Accordingly, high BDE-47/-99 and low BDE-99/-100 ratios indicate that BDE99 photo-degrades during LRAT. In the GLB air samples, only high ratios of BDE-47/-99 were observed. BDE-47 is a more volatile compound than BDE-99[4]; our results (high BDE-47/-99 ratios) indicate that BDE-47 must be coming from secondary emissions (e.g. from soils) or primary emissions (e.g. from BDE treated products).

The *anti*-DDC-CO fractional abundance (f_{anti}) can be used to assess the fate and distribution of DDC-CO in the environment. The f_{anti} was calculated as the ratio of the concentration of the *anti*-DDC-CO isomer divided by the sum of the concentrations of the *anti*- plus *syn*-DDC-CO isomers in the gas plus particle phase. Commercial DDC-CO product from OxyChem contains about 75 % *anti*-DDC-CO and 25 % *syn*-DP, giving a f_{anti} value of 0.75[5]. The annual average f_{anti} values at Point Petre and Burnt Island are listed in Table S5. The values f_{anti} range from 0.67 to 0.84, which are similar to that for the commercial product. This suggests that both DDC-CO isomers have similar atmospheric persistence and that no obvious stereo-selective process has occurred during atmospheric transport. This is consistent with the study by Venier and Hites[6], who reported a similar fate for these two isomers in the atmosphere. However, Salamova et al.[7] observed a decrease in f_{anti} in particle phase with increasing distance from the source area, indicating that *syn*-DDC-CO is more stable toward photo-degradation during LRAT.

Section S3. Gas-Particle Partitioning of HFRs

The fate, transport and removal of semi-volatile organic compounds (SVOCs) from the atmosphere via dry and wet deposition are influenced by their gas-particle partitioning. The degree of particle sorption is related to the vapor pressure of a chemical, e.g. low volatility compounds such as BDE-209 and DBDPE present mainly in particulate phase. The *G/P* partitioning behavior of SVOCs is defined either by the particle/gas partition coefficient, K_P (m^3 air $\cdot ug^{-1}$ of particles) or the fraction of the total concentration of a SVOC in the atmosphere sorbed to particles, ϕ [8]. The ϕ correlates strongly with sub-cooled liquid vapor pressure, P_L (Pa). One- or two- parameter non-linear model can be used to regress between ϕ and P_L. Su et al.[8] suggested that regression between ϕ and P_L by the one- and two-parameter non-linear models are comparable, thus the one-parameter non-linear model (Junge-Pankow model)[9] as show in Eq.1 was used to fit gas/particle partitioning of PBDEs from different sampling events at Point Petre in this study.

$$\phi = A/ (A + P_L) \quad (1)$$

where A is a fitting parameter. The final adjusted P_L values at 25 °C for BDE-28, BDE-47, BDE-99, BDE-100 and BDE-153 are reported in Wania and Dugani[4] but are not available for BDE-17, BDE-49, BDE-66, BDE-85, BDE-138, BDE-154, BDE-183 and BDE-209. Thus, their P_L values at 25 °C were derived from the correlation between P_L and Le Bas Molar volume[4]. The P_L values for the PBDEs at the ambient temperature of the sampling period at Point Petre were calculated using final adjusted P_L and ΔH_{vap} data[10].

The one-parameter non-linear regression was performed for 114 sampling events, where each sampling event contained 6 or more pairs of ϕ and P_L. The regression results are reported in Table S6. The correlation coefficient, R^2, ranged between 0.40 and 0.99, except for 15 sampling events. The fitting parameter A ranged from 1.5 x 10^{-9} to 3.6 x 10^{-5}. A is related to partitioning capacity of particles for SVOCs. Adsorption onto the particle surface[11] and absorption into the organic matter of the particles[12] are two mechanisms, which have been used to explain the gas/particle partitioning behavior of SVOCs. In the case of adsorptive mechanism, A is related to particle surface area θ, whereas in the case of absorption mechanism, A is determined from the concentrations of the total suspended particles *TSP* and organic matter content of particles f_{OM}. The θ, *TSP* and f_{OM} can vary widely in background, urban and rural air[8]. We observed a large variability in the A values in this study which is likely due to the different sorptive properties of the particles as a result of their different origins to the sampling sites and also the *TSP*.

Table S1. Annual mean and standard deviation for field and laboratory blank

Part1. Burnt Island from 2005 to 2013

	Field Blank **GFF** (Average ± SD)	**Field blank** **PUF** (Average ± SD)	**Lab blank** (Average ± SD)
Non-PBDE HFRs			
***anti*-DDC-CO**			
2008			
2009		0.024 ± 0.033	
2010			
2011			
2012			0.0048 ± 0.0118
2013			
***syn*-DDC-CO**			
2008			
2009			0.0048 ± 0.012
2010			
2011			0.0033 ± 0.0082
2012			
2013			
TBP-AE			
2009		0.059 ± 0.042	
2010		0.047 ± 0.034	0.0059 ± 0.0088
2011		0.095 ± 0.084	0.010 ± 0.022
2012	0.012 ± 0.016	0.057 ± 0.027	0.0076 ± 0.017
2013	0.012 ± 0.016		
TBP-BAE			
2009			
2010			
2011			
2012			
2013			
BTBPE			
2009			
2010			0.0051 ± 0.018
2011			0.010 ± 0.025
2012			
2013			
DBDPE			
2009			
2010			
2011			

	Field Blank **GFF** (Average ± SD)	**Field blank** **PUF** (Average ± SD)	**Lab blank** (Average ± SD)
2012	0.021 ± 0.042		
2013			
DPTE			
2009			
2010			
2011		0.018 ± 0.012	
2012			
2013			
HBBz			
2009		0.024 ± 0.033	
2010	0.042 ± 0.016	0.094 ± 0.023	0.040 ± 0.029
2011		0.018 ± 0.012	0.0049 ± 0.012
2012		0.019 ± 0.022	
2013			
HBCDD			
2009			
2010			
2011			
2012			
2013			
PBEB			
2009			
2010			
2011		0.0012 ± 0.0024	
2012			
2013			
PBT			
2009			
2010	0.024 ± 0.013	0.046 ± 0.0072	0.028 ± 0.016
2011		0.0058 ± 0.011	0.0035 ± 0.0085
2012		0.0074 ± 0.0086	0.0040 ± 0.0070
2013			
EH-TBB			
2009			
2010			0.043 ± 0.15
2011			
2012			
2013			
PBDEs			
BDE-17			
2005			

	Field Blank **GFF** (Average ± SD)	**Field blank** **PUF** (Average ± SD)	**Lab blank** (Average ± SD)
2006			
2007			
2008			
2009			
2010			
2011			
2012			
2013			
BDE-28			
2005			
2006			
2007			
2008			
2009			
2010			
2011		0.013 ± 0.026	
2012		0.020 ± 0.017	
2013			
BDE-47			
2005		1.2 ± 0.14	0.25 ± 0.26
2006		1.1 ± 0.073	0.18 ± 0.22
2007		0.68 ± 0.29	0.090 ± 0.17
2008		0.93 ± 0.57	0.021 ± 0.069
2009		0.68 ± 0.28	0.0839 ± 0.11
2010		0.55 ± 0.092	0.12 ± 0.12
2011		0.22 ± 0.25	0.082 ± 0.072
2012		0.68 ± 0.094	0.31 ± 0.23
2013		0.33 ± 0.00	
BDE-49			
2005			
2006			
2007			
2008			
2009			
2010		0.013 ± 0.018	
2011		0.011 ± 0.022	
2012			
2013			
BDE-66			
2005			
2006			
2007			
2008			

	Field Blank **GFF** (Average ± SD)	**Field blank** **PUF** (Average ± SD)	**Lab blank** (Average ± SD)
2009			
2010			
2011			
2012			
2013			
BDE-71			
2005			
2006			
2007			
2008			
2009			
2010			
2011			
2012			
2013			
BDE-85			
2005			
2006			
2007			
2008			
2009			
2010			
2011			
2012			
2013			
BDE-99			
2005		1.1 ± 0.38	0.15 ± 0.20
2006		1.2 ± 0.44	0.20 ± 0.22
2007		0.39± 0.17	
2008		0.77 ± 0.56	
2009		0.27 ± 0.20	0.069 ± 0.14
2010		0.54 ± 0.16	0.15 ± 0.16
2011		0.34 ± 0.35	0.094 ± 0.11
2012		0.25 ± 0.067	0.16 ± 0.11
2013		0.41 ± 0.00	
BDE-100			
2005		0.25 ± 0.15	
2006		0.23 ± 0.15	
2007			
2008			
2009		0.089 ± 0.034	0.014 ± 0.035
2010		0.13 ± 0.041	0.037 ± 0.042
2011		0.066 ± 0.068	0.024 ± 0.026
2012		0.075 ± 0.016	0.054 ± 0.048

	Field Blank **GFF** (Average ± SD)	**Field blank** **PUF** (Average ± SD)	**Lab blank** (Average ± SD)
2013		0.11 ± 0.00	
BDE-138			
2005			
2006			
2007			
2008			
2009			
2010			
2011			
2012			
2013			
BDE-153			
2005			
2006			
2007			
2008			
2009			
2010			
2011			
2012			
2013			
BDE-154			
2005			
2006			
2007			
2008			
2009		0.070 ± 0.091	
2010		0.022 ± 0.025	0.0019 ± 0.0065
2011			0.0038 ± 0.0091
2012			
2013			
BDE-183			
2005			
2006			
2007			
2008			
2009			
2010			
2011			
2012			
2013			
BDE-209			
2005			

	Field Blank **GFF** (Average ± SD)	**Field blank** **PUF** (Average ± SD)	**Lab blank** (Average ± SD)
2006			0.0020 ± 0.0076
2007		0.27 ± 0.42	0.021 ± 0.073
2008		0.24 ± 0.38	
2009		0.071 ± 0.099	
2010			
2011			
2012			
2013			0.0020 ± 0.0076

Part2. Egbert from 2005 to 2008

	Field Blank **GFF** (Average ± SD)	**Field blank** **PUF** (Average ± SD)	**Lab blank** (Average ± SD)
Non-PBDE HFRs			
anti-DDC-CO			
2008			
syn-DDC-CO			
2008			
PBDEs			
BDE-17			
2005			
2006			
2007			
2008			
BDE-28			
2005			
2006			
2007			
2008			
BDE-47			
2005	0.46 ± 0.16	1.4 ± 0.66	0.17 ± 0.20
2006	0.28 ± 0.27	1.2 ± 0.52	0.18 ± 0.19
2007	0.15 ± 0.13	0.75 ± 0.14	0.11 ± 0.20
2008		0.79 ± 0.00	
BDE-49			
2005			
2006			
2007			
2008			
BDE-66			
2005			
2006			
2007			
2008			
BDE-71			
2005			
2006			
2007			
2007			
2008			

	Field Blank **GFF** (Average ± SD)	**Field blank** **PUF** (Average ± SD)	**Lab blank** (Average ± SD)
BDE-85			
2005			
2006			
2007			
2008			
BDE-99			
2005	0.49 ± 0.18	0.74 ± 0.16	0.15 ± 0.19
2006	0.29 ± 0.22	1.1 ± 0.45	0.14 ± 0.22
2007	0.080 ± 0.096	0.54 ± 0.10	0.062 ± 0.14
2008		0.74 ± 0.00	
BDE-100			
2005	0.071 ± 0.073	0.18 ± 0.18	
2006		0.26 ± 0.16	
2007			
2008		0.23 ± 0.00	
BDE-138			
2005			
2006			
2007			
2008			
BDE-153			
2005			
2006			
2007			
2008			
BDE-154			
2005			
2006			
2007			
2008			
BDE-183			
2005			
2006			
2007			
2008			
BDE-209			
2005	0.070 ± 0.11		
2006	0.14 ± 0.25		0.029 ± 0.10
2007	0.053 ± 0.063		
2008			

204

Part 3. Point Petre from 2005 to 2014

	Field Blank **GFF** (Average ± SD)	**Field blank** **PUF** (Average ± SD)	**Lab blank** (Average ± SD)
Non-PBDE HFRs			
***anti*-DDC-CO**			
2008	0.051 ± 0.076	0.047 ± 0.049	
2009			0.0044 ± 0.016
2010			0.0016 ± 0.0037
2011			
2012	0.0056 ± 0.0087		
2013			0.0070 ± 0.022
2014			0.0063 ± 0.021
***syn*-DDC-CO**			
2008	0.034 ± 0.047		
2009			0.0066 ± 0.024
2010			0.0033 ± 0.0078
2011			
2012			
2013			
2014			
TBP-AE			
2009		0.0471 ± 0.026	0.0071 ± 0.0098
2010	0.018 ± 0.016	0.048 ± 0.059	0.0092 ± 0.021
2011		0.031 ± 0.035	0.00081 ± 0.0031
2012		0.048 ± 0.014	0.0087 ± 0.015
2013			
2014		0.046 ± 0.046	
TBP-BAE			
2009			
2010			
2011			
2012			
2013			
2014			
BTBPE			
2009			
2010			0.0094 ± 0.022
2011			0.0034 ± 0.013
2012			
2013			
2014			
DBDPE			
2009			

	Field Blank **GFF** (Average ± SD)	**Field blank** **PUF** (Average ± SD)	**Lab blank** (Average ± SD)
2010			
2011			
2012			
2013			
2014			
DPTE			
2009			
2010			
2011			0.0016 ± 0.0063
2012			
2013			
2014		0.12 ± 0.11	
HBBz			
2009		0.015 ± 0.029	0.023 ± 0.025
2010	0.023 ± 0.013	0.048 ± 0.036	0.0057 ± 0.013
2011			0.0014 ± 0.0055
2012		0.013 ± 0.020	0.0012 ± 0.0040
2013			
2014			
HBCDD			
2009			
2010			
2011			
2012			
2013			
2014			
PBEB			
2009			
2010			
2011			
2012			
2013			
2014			
PBT			
2009			0.019 ± 0.019
2010	0.023 ± 0.013		0.0043 ± 0.010
2011			
2012			
2013			
2014			
EH-TBB			
2009			

	Field Blank **GFF** (Average ± SD)	**Field blank** **PUF** (Average ± SD)	**Lab blank** (Average ± SD)
2010			
2011			
2012			0.011 ± 0.037
2013			
2014			
PBDEs			
BDE-17			
2005			
2006			
2007			
2008			
2009			
2010			
2011			
2012			
2013			
2014			
BDE-28			
2005			
2006			
2007			
2008			
2009	0.012 ± 0.016		
2010			
2011			
2012		0.017 ± 0.011	
2013			
2014			
BDE-47			
2005	0.38 ± 0.074	1.48 ± 0.57	0.16 ± 0.22
2006	0.28 ± 0.11	1.4 ± 0.59	0.17 ± 0.20
2007	0.20 ± 0.20	0.56 ± 0.22	0.12 ± 0.17
2008	0.47 ± 0.043	0.74 ± 0.20	0.25 ± 0.17
2009	0.27 ± 0.20	0.49 ± 0.15	0.11 ± 0.11
2010	0.070 ± 0.039	0.43 ± 0.31	0.13 ± 0.010
2011	0.088 ± 0.013	0.49 ± 0.20	0.11 ± 0.15
2012	0.047 ± 0.046	0.18 ± 0.20	0.064 ± 0.094
2013	0.061 ± 0.042	0.37 ± 0.030	0.054 ± 0.071
2014	0.31 ± 0.12	0.90 ± 0.39	0.73 ± 0.49
BDE-49			
2005			
2006			

	Field Blank **GFF** (Average ± SD)	**Field blank** **PUF** (Average ± SD)	**Lab blank** (Average ± SD)
2007			
2008			
2009			
2010			
2011			
2012			
2013			
2014			
BDE-66			
2005			
2006			
2007			0.0022 ± 0.0079
2008			
2009			
2010			
2011			
2012			
2013			
2014			
BDE-71			
2005			
2006			
2007			
2008			
2009			
2010			
2011			
2012			
2013			
2014			
BDE-85			
2005			
2006			
2007			0.0022 ± 0.0079
2008			0.016 ± 0.061
2009			0.0022 ± 0.0079
2010			
2011			0.00054 ± 0.0021
2012			
2013			
2014			
BDE-99			
2005	0.33 ± 0.084	1.1 ± 0.63	0.073 ± 0.14
2006	0.19 ± 0.10	0.97 ± 0.34	0.13 ± 0.19

	Field Blank **GFF** (Average ± SD)	**Field blank** **PUF** (Average ± SD)	**Lab blank** (Average ± SD)
2007		0.27 ± 0.25	0.046 ± 0.087
2008	0.32 ± 0.039	0.58 ± 0.18	0.37 ± 0.76
2009	0.12 ± 0.062	0.52 ± 0.31	0.12 ± 0.13
2010	0.05 ± 0.032	0.39 ± 0.32	0.15 ± 0.13
2011	0.071 ± 0.020	0.46 ± 0.15	0.10 ± 0.18
2012	0.034 ± 0.034	0.19 ± 0.23	0.077 ± 0.084
2013	0.026 ± 0.051	0.36 ± 0.17	0.063 ± 0.093
2014	0.12 ± 0.062	0.163 ± 0.19	0.13 ± 0.15
BDE-100			
2005		0.21 ± 0.20	0.011 ± 0.032
2006		0.18 ± 0.18	0.0088 ± 0.021
2007			0.013 ± 0.022
2008	0.084 ± 0	0.15 ± 0.034	0.079 ± 0.12
2009	0.035 ± 0.025	0.13 ± 0.077	0.029 ± 0.031
2010	0.012 ± 0.016	0.15 ± 0.15	0.044 ± 0.046
2011		0.11 ± 0.037	0.014 ± 0.040
2012	0.012 ± 0.0030	0.12 ± 0.12	0.0030 ± 0.011
2013			
2014	0.020 ± 0.029	0.13 ± 0.12	0.022 ± 0.050
BDE-138			
2005			
2006			
2007			0.0044 ± 0.016
2008			0.0041 ± 0.015
2009			0.0044 ± 0.016
2010			
2011			
2012			
2013			
2014			
BDE-153			
2005			
2006			
2007			0.0044 ± 0.016
2008			0.029 ± 0.11
2009			0.0044 ± 0.016
2010			
2011			
2012			
2013			
2014			
BDE-154			
2005			
2006			

	Field Blank **GFF** (Average ± SD)	**Field blank** **PUF** (Average ± SD)	**Lab blank** (Average ± SD)
2007			0.0022 ± 0.0079
2008		0.024 ± 0.032	0.024 ± 0.092
2009		0.024 ± 0.032	0.0022 ± 0.0079
2010		0.053 ± 0.083	0.0019 ± 0.0064
2011			
2012		0.019 ± 0.025	
2013			
2014			
BDE-183			
2005			
2006			0.075± 0.27
2007			0.33 ± 1.2
2008			1.3 ± 3.4
2009			0.40 ± 1.2
2010			
2011			
2012			
2013			
2014			
BDE-209			
2005	0.012 ± 0.024		
2006	0.050 ± 0.062	0.098 ± 0.12	
2007	0.082 ± 0.12		0.022 ± 0.070
2008		1.6 ± 2.1	
2009			
2010			
2011			
2012			
2013			
2014			

TableS2. The % breakthrough for very volatile compounds

	BDE-17	BDE-28	BDE-47	BDE-49	TBP-AE	TBP-BAE	PBT	TBP-DBPE	PBEB	HBBz
Break Through (%)	0	3-12	7-30	7-15	5-38	0	0	0	20-25	7-21

Table S3. Concentrations of halogenated flame retardants at three sampling sites in the Canadian Great Lakes Basin. When there is no data showing, it means the HFR compound was not detected.

Part 1. Burnt Island from 2005 to 2013

	Total (GFF+PUF) Mean (STD; Min-Max)	Gas Phase Mean (STD; Min-Max)	Particle Phase Mean (STD; Min-Max)
Non-PBDE HFRs			
***anti*-DDC-CO**			
2008	3.2 (4.6; 0.12 - 12)		3.2 (4.6; 0.12 - 12)
2009	0.24 (0.35; 0.030 - 1.3)	0.060 (0.0024; 0.055 - 0.062)	0.24 (0.35; 0.030 - 1.3)
2010	0.27 (0.41; 0.029 - 1.8)	0.083 (0.050; 0.046 - 0.14)	0.27 (0.42; 0.029 - 1.8)
2011	1.0 (2.2; 0.053 - 5.5)	0.11	1.0 (2.2; 0.053 - 5.5)
2012	0.34 (0.50; 0.026 - 1.8)		0.34 (0.50; 0.026 - 1.8)
2013	0.80		0.80
***syn*-DDC-CO**			
2008	2.4 (3.6; 0.23 - 7.8)		2.4 (3.6; 0.23 - 7.8)
2009	0.090 (0.10; 0.028 - 0.44)	0.11 (0.080; 0.055 - 0.24)	0.074 (0.083; 0.028 - 0.37)
2010	0.13 (0.23; 0.029 - 0.91)		0.13 (0.23; 0.029 - 0.91)
2011	0.30 (0.40; 0.066 - 1.1)	0.092	0.29 (0.45; 0.066 - 1.1)
2012	0.27 (0.36; 0.018 - 1.0)		0.27 (0.36; 0.018 - 0.98)
2013	0.22		0.22
TBP-AE			
2009	0.17 (0.12; 0.028 - 0.48)	0.16 (0.11; 0.059 - 0.48)	0.036 (0.012; 0.028 - 0.061)
2010	0.31 (0.15; 0.083 - 0.60)	0.30 (0.15; 0.083 - 0.57)	0.030 (0.00079; 0.029 - 0.031)
2011	0.23 (0.20; 0.047 - 0.86)	0.22 (0.20; 0.047 - 0.86)	0.048 (0.043; 0.018 - 0.079)
2012	0.17 (0.14; 0.061 - 0.63)	0.16 (0.14; 0.061 - 0.63)	
2013	0.085	0.085	0.11
TBP-BAE			
2009	0.087 (0.057; 0.060 - 0.19)	0.087 (0.057; 0.060 - 0.19)	
2010	0.083 (0.021; 0.059 - 0.10)	0.083 (0.021; 0.059 - 0.10)	
2011	0.039 (0.015; 0.019 - 0.062)	0.039 (0.015; 0.019 - 0.062)	
2012	0.025 (0.023; 0.0047 - 0.052)	0.045 (0.010; 0.038 - 0.052)	0.0057 (0.0015; 0.0047 - 0.0068)
2013			
BTBPE			
2009	0.081 (0.074; 0.029 - 0.28)		0.081 (0.074; 0.029 - 0.28)
2010	0.14 (0.14; 0.030 - 0.47)	0.12	0.15 (0.15; 0.030 - 0.47)
2011	0.42	0.42	
2012	0.044	0.044	
2013			
DBDPE			
2009			
2010			
2011	1.2		1.2
2012	0.59 (0.22; 0.27 - 0.88)		0.59 (0.22; 0.27 - 0.88)

	Total (GFF+PUF) Mean (STD; Min-Max)	Gas Phase Mean (STD; Min-Max)	Particle Phase Mean (STD; Min-Max)
2013			
DPTE			
2009	0.050 (0.017; 0.031 - 0.060)	0.060 (0.00037; 0.060 - 0.060)	
2010	0.10 (0.071; 0.029 - 0.21)	0.12 (0.058; 0.037 - 0.18)	0.036 (0.013; 0.029 - 0.059)
2011	0.043 (0.026; 0.023 - 0.11)	0.032 (0.013; 0.020 - 0.055)	0.069 (0.054; 0.031 - 0.11)
2012	0.027 (0.013; 0.013 - 0.040)	0.034	0.024 (0.014; 0.013 - 0.040)
2013	0.11 (0.019; 0.091 - 0.13)	0.11 (0.019; 0.091 - 0.13)	
HBBz			
2009	0.18 (0.18; 0.029 - 0.95)	0.14 (0.18; 0.055 - 0.92)	0.067 (0.074; 0.029 - 0.37)
2010	0.30 (0.19; 0.059 - 0.84)	0.23 (0.15; 0.073 - 0.72)	0.087 (0.058; 0.029 - 0.33)
2011	0.14 (0.093; 0.048 - 0.37)	0.12 (0.091; 0.034 - 0.32)	0.046 (0.043; 0.011 - 0.13)
2012	0.17 (0.078; 0.060 - 0.31)	0.14 (0.079; 0.042 - 0.28)	0.031 (0.013; 0.014 - 0.059)
2013			
HBCDD			
2009	0.17 (0.27; 0.030 - 0.92)	0.19	0.17 (0.28; 0.030 - 0.92)
2010	0.50 (0.81; 0.091 - 2.0)		0.50 (0.81; 0.091 - 2.0)
2011			
2012	0.26 (0.30; 0.039 - 0.80)		0.26 (0.30; 0.039 - 0.80)
2013			
PBEB			
2009	0.060 (0.0015; 0.059 - 0.062)	0.061 (0.0016; 0.060 - 0.062)	0.059
2010	0.051 (0.029; 0.019 - 0.12)	0.051 (0.029; 0.019 - 0.12)	
2011	0.021 (0.011; 0.0078 - 0.041)	0.021 (0.011; 0.0078 - 0.041)	
2012	0.018 (0.010; 0.0031 - 0.028)	0.023 (0.0046; 0.018 - 0.028)	0.0031 (0.000093; 0.0030 - 0.0032)
2013			
PBT			
2009		0.061 (0.0020; 0.057 - 0.063)	
2010		0.077 (0.025; 0.047 - 0.16)	
2011		0.059 (0.084; 0.0075 - 0.27)	
2012		0.041 (0.026; 0.014 - 0.12)	
2013			
EH-TBB			
2009	0.72 (0.42; 0.30 - 1.5)	0.52 (0.41; 0.060 - 1.5)	0.66 (0.46; 0.20 - 1.4)
2010	4.1 (6.0; 0.17 - 20)	6.2 (6.7; 0.54 - 20)	0.33 (0.10; 0.17 - 0.45)
2011	1.0 (1.0; 0.12 - 3.4)	0.94 (1.1; 0.12 - 3.2)	0.40 (0.22; 0.19 - 0.67)
2012	1.0 (2.0; 0.042 - 5.6)	3.4 (3.0; 1.2 - 5.5)	0.18 (0.24; 0.042 - 0.76)
2013			
PBDEs			
BDE-17			
2005	0.39 (0.17; 0.15 - 0.7)	0.39 (0.17; 0.15 - 0.74)	
2006	0.31 (0.14; 0.15 - 0.46)	0.31 (0.14; 0.15 - 0.46)	
2007	0.47 (0.32; 0.24 - 1.0)	0.50 (0.36; 0.24 - 1.0)	0.36

	Total (GFF+PUF) Mean (STD; Min-Max)	Gas Phase Mean (STD; Min-Max)	Particle Phase Mean (STD; Min-Max)
2008	0.42 (0.27; 0.18 - 1.1)	0.42 (0.27; 0.18 - 1.1)	
2009	0.19 (0.19; 0.028 - 0.84)	0.19 (0.20; 0.057 - 0.84)	0.039 (0.021; 0.028 - 0.089)
2010	0.41 (1.1; 0.028 - 5.6)	0.45 (1.2; 0.032 - 5.6)	0.029 (0.00070; 0.028 - 0.030)
2011	0.11 (0.080; 0.016 - 0.24)	0.12 (0.084; 0.016 - 0.24)	0.073 (0.072; 0.027 - 0.16)
2012	0.20 (0.22; 0.0062 - 0.82)	0.17 (0.13; 0.043 - 0.47)	0.083 (0.20; 0.0037 - 0.57)
2013			
BDE-28			
2005	0.64 (0.45; 0.13 - 1.9)	0.64 (0.45; 0.13 - 1.9)	
2006	0.48 (0.39; 0.093 - 1.3)	0.48 (0.39; 0.093 - 1.3)	
2007	0.76 (0.74; 0.12 - 2.9)	0.71 (0.71; 0.12 - 2.7)	0.70 (0.73; 0.19 - 1.2)
2008	0.92 (0.71; 0.093 - 2.8)	1.0 (0.70; 0.30 - 2.8)	
2009	0.39 (0.52; 0.030 - 2.5)	0.38 (0.52; 0.057 - 2.5)	0.057 (0.054; 0.028 - 0.24)
2010	0.42 (0.68; 0.030 - 3.4)	0.42 (0.68; 0.027 - 3.3)	0.043 (0.028; 0.028 - 0.14)
2011	0.28 (0.17; 0.046 - 0.51)	0.23 (0.18; 0.020 - 0.51)	0.13 (0.17; 0.036 - 0.49)
2012	0.43 (0.35; 0.098 - 1.1)	0.37 (0.29; 0.048 - 0.98)	0.067 (0.14; 0.0047 - 0.51)
2013	0.030		0.030
BDE-47			
2005	7.7 (6.7; 1.6 - 26)	6.5 (5.8; 0.95 - 18)	1.3 (2.0; 0.34 - 12)
2006	6.7 (6.2; 1.3 - 31)	5.3 (5.1; 0.72 - 22)	1.3 (1.9; 0.24 - 8.8)
2007	17 (54; 0.60 - 290)	4.8 (7.1; 0.40 - 36)	12 (53; 0.17 - 290)
2008	17 (32; 0.14 - 170)	16 (34; 0.69 - 170)	3.0 (8.0; 0.11 - 36)
2009	11 (14; 0.82 - 54)	6.3 (10; 0.58 - 53)	4.7 (10; 0.18 - 37)
2010	10 (14; 0.90 - 48)	6.4 (10; 0.78 - 38)	3.8 (9.4; 0.088 - 40)
2011	20 (34; 1.8 - 140)	4.8 (4.7; 0.55 - 14)	15 (36; 0.27 - 140)
2012	7.5 (6.1; 1.9 - 24)	4.5 (3.0; 0.87 - 11)	3.2 (6.0; 0.35 - 23)
2013	6.9 (6.9; 0.74 - 14)	3.3 (5.6; 0.35 - 12)	4.8 (7.4; 0.24 - 13)
BDE-49			
2005	0.46 (0.25; 0.084 - 0.99)	0.47 (0.21; 0.16 - 0.99)	0.19 (0.15; 0.084 - 0.29)
2006	0.42 (0.31; 0.14 - 1.0)	0.42 (0.31; 0.14 - 1.0)	
2007	1.5 (1.9; 0.29 - 4.9)	0.60 (0.40; 0.29 - 1.3)	3.0 (2.7; 1.1 - 4.9)
2008	0.90 (0.97; 0.12 - 3.9)	0.91 (1.1; 0.12 - 3.9)	0.65 (0.08; 0.59 - 0.71)
2009	0.34 (0.38; 0.030 - 1.6)	0.26 (0.34; 0.057 - 1.6)	0.15 (0.24; 0.030 - 0.86)
2010	0.33 (0.39; 0.029 - 1.4)	0.25 (0.32; 0.030 - 1.3)	0.11 (0.21; 0.028 - 0.83)
2011	0.51 (0.79; 0.056 - 3.2)	0.18 (0.16; 0.029 - 0.47)	0.65 (1.1; 0.032 - 3.2)
2012	0.27 (0.24; 0.012 - 0.94)	0.19 (0.10; 0.063 - 0.39)	0.12 (0.19; 0.011 - 0.57)
2013	0.22 (0.16; 0.043 - 0.34)	0.29	0.19 (0.21; 0.043 - 0.34)
BDE-66			
2005	0.50 (0.33; 0.076 - 0.99)	0.47 (0.19; 0.26 - 0.71)	0.20 (0.11; 0.076 - 0.28)
2006	0.42 (0.11; 0.32 - 0.57)	0.42 (0.11; 0.32 - 0.57)	
2007	3.4 (2.6; 1.6 - 5.2)	0.56	3.1 (3.0; 1.0 - 5.2)
2008	1.0 (1.30; 0.24 - 3.6)	1.2 (1.6; 0.24 - 3.6)	0.61 (0.060; 0.56 - 0.65)
2009	0.33 (0.29; 0.030 - 0.90)	0.30 (0.24; 0.058 - 0.84)	0.22 (0.27; 0.030 - 0.77)
2010	0.26 (0.29; 0.028 - 0.95)	0.23 (0.23; 0.057 - 0.76)	0.15 (0.23; 0.028 - 0.83)
2011	0.39 (0.75; 0.026 - 3.0)	0.11 (0.085; 0.018 - 0.26)	0.67 (1.1; 0.026 - 3.0)

	Total (GFF+PUF) Mean (STD; Min-Max)	Gas Phase Mean (STD; Min-Max)	Particle Phase Mean (STD; Min-Max)
2012	0.15 (0.21; 0.0090 - 0.75)	0.091 (0.045; 0.044 - 0.17)	0.12 (0.19; 0.0090 - 0.58)
2013	0.20 (0.092; 0.14 - 0.27)	0.14	0.27
BDE-71			
2005			
2006			
2007	0.44		0.44
2008			
2009	0.078 (0.053; 0.029 - 0.18)	0.12 (0.086; 0.058 - 0.18)	0.058 (0.022; 0.029 - 0.083)
2010	0.15 (0.16; 0.040 - 0.33)		0.064 (0.035; 0.040 - 0.089)
2011	0.11 (0.11; 0.049 - 0.31)	0.059 (0.014; 0.049 - 0.068)	0.15 (0.14; 0.054 - 0.31)
2012	0.54		0.54
2013			
BDE-85			
2005	0.64 (0.52; 0.27 - 1.0)	0.27	1.0
2006	0.45 (0.083; 0.36 - 0.52)		0.45 (0.083; 0.36 - 0.52)
2007	8.4 (12; 0.39 - 26)		8.4 (12; 0.39 - 26)
2008	4.8 (5.9; 0.23 - 13)	13	1.9 (1.5; 0.23 -3.0_
2009	0.58 (1.05; 0.028 - 3.2)	0.31 (0.80; 0.058 - 3.0)	0.44 (0.95; 0.028 - 3.2)
2010	0.70 (1.3; 0.030 - 5.1)	0.26 (0.67; 0.057 - 2.9)	0.73 (1.4; 0.030 - 5.1)
2011	1.9 (4.0; 0.044 - 14)	0.20 (0.21; 0.044 - 0.53)	5.0 (5.9; 0.80 - 14)
2012	0.29 (0.58; 0.0090 - 1.9)	0.038 (0.0079; 0.032 - 0.043)	0.29 (0.57; 0.0090 - 1.9)
2013	1.0 (0.060; 1.0 - 1.1)	1.1	1.0
BDE-99			
2005	3.7 (4.2; 1.0 - 23)	2.3 (1.8; 0.65 - 7.0)	1.4 (3.6; 0.36 - 20)
2006	4.1 (3.8; 1.3 - 17)	2.4 (1.6; 0.55 - 6.1)	1.8 (3.3; 0.22 - 15)
2007	22 (92; 1.0 - 500)	0.98 (0.81; 0.35 - 4.2)	22 (93; 0.14 - 500)
2008	17 (53; 0.2 - 290)	14 (56; 0.40 - 290)	4.6 (15; 0.11 - 62)
2009	11 (20; 0.65 - 66)	3.1 (9.5; 0.30 - 53)	7.6 (18; 0.12 - 66)
2010	11 (24; 0.58 - 110)	3.7 (11; 0.44 - 60)	7.9 (22; 0.088 - 100)
2011	27 (66; 1.1 - 260)	1.3 (0.87; 0.57 - 3.4)	26 (67; 0.24 - 260)
2012	5.3 (10; 0.72 - 40)	0.91 (0.52; 0.28 - 2.0)	4.7 (11; 0.29 - 40)
2013	11 (12; 0.58 - 21)	5.6 (10; 0.42 - 21)	7.3 (12; 0.29 - 21)
BDE-100			
2005	0.95 (0.94; 0.080 - 4.9)	0.76 (0.50; 0.14 - 1.9)	0.30 (0.65; 0.079 - 3.7)
2006	0.93 (0.81; 0.23 - 3.7)	0.63 (0.40; 0.13 - 1.6)	0.49 (0.70; 0.12 - 2.8)
2007	6.1 (20; 0.084 - 91)	0.51 (0.41; 0.090 - 1.6)	9.6 (26; 0.084 - 91)
2008	4.5 (12; 0.12 - 55)	3.8 (12; 0.12 - 55)	1.6 (4.0; 0.12 - 12)
2009	2.2 (3.7; 0.14 - 12)	0.74 (1.8; 0.059 - 9.5)	1.5 (3.4; 0.028 - 12)
2010	2.2 (4.3; 0.13 - 18)	0.85 (2.1; 0.10 - 11)	1.4 (3.7; 0.029 - 18)
2011	5.4 (12; 0.25 - 48)	0.41 (0.35; 0.12 - 1.2)	5.0 (13; 0.050 - 48)
2012	1.0 (2.1; 0.24 - 8.0)	0.33 (0.21; 0.086 - 0.78)	1.0 (2.2; 0.066 - 8.0)
2013	2.2 (2.3; 0.15 - 4.2)	1.4 (2.3; 0.081 - 4.1)	1.4 (2.3; 0.061 - 4.1)

	Total (GFF+PUF) Mean (STD; Min-Max)	Gas Phase Mean (STD; Min-Max)	Particle Phase Mean (STD; Min-Max)
BDE-138			
2005			
2006			
2007	2.5 (2.9; 0.35 - 5.8)		2.5 (2.9; 0.35 - 5.8)
2008	1.3 (1.2; 0.59 - 2.7)	2.68	0.65 (0.083; 0.59 - 0.71)
2009	0.34 (0.31; 0.030 - 0.73)	0.73	0.30 (0.29; 0.030 - 0.71)
2010	0.55 (0.39; 0.090 - 1.2)	0.62	0.45 (0.42; 0.090 - 1.2)
2011	1.1 (1.3; 0.29 - 2.5)		1.1 (1.3; 0.29 - 2.5)
2012	0.31 (0.14; 0.15 - 0.42)		0.31 (0.14; 0.15 - 0.42)
2013	0.27 (0.072; 0.22 - 0.32)	0.32	0.22
BDE-153			
2005	0.88 (0.97; 0.20 - 2.0)		1.1 (1.3; 0.20 - 2.0)
2006	0.88 (0.30; 0.67 - 1.2)		0.88 (0.30; 0.67 - 1.2)
2007	16 (22; 0.87 - 49)		16 (22; 0.87 - 49)
2008	7.5 (8.9; 0.52 - 23)	13 (14; 2.8 - 23)	4.0 (3.0; 0.52 - 6.1)
2009	0.95 (2.0; 0.028 - 6.3)	0.55 (1.6; 0.055 - 6.0)	0.72 (1.7; 0.028 - 6.3)
2010	1.2 (2.5; 0.029 - 10)	0.38 (1.2; 0.037 - 5.3)	1.1 (2.6; 0.029 - 10)
2011	5.6 (9.4; 0.047 - 24)		5.6 (9.4; 0.047 - 24)
2012	0.48 (1.1; 0.011 - 3.7)	0.099	0.47 (1.1; 0.010 - 3.7)
2013	1.4 (1.1; 0.068 - 2.1)	2.1	1.0 (1.4; 0.068 - 2.0)
BDE-154			
2005	1.1 (0.75; 0.54 - 1.6)		1.1 (0.75; 0.54 - 1.6)
2006	0.69 (0.36; 0.24 - 1.2)		0.72 (0.41; 0.24 - 1.2)
2007	14 (19; 0.75 - 41)		14 (19; 0.75 - 41)
2008	7.7 (9.6; 0.43 - 25)	14 (15; 3.0 - 25)	3.6 (2.8; 0.43 - 5.6)
2009	0.89 (1.7; 0.029 - 5.4)	0.39 (1.2; 0.055 - 5.3)	0.69 (1.6; 0.028 - 5.2)
2010	1.0 (2.0; 0.029 - 8.4)	0.28 (1.0; 0.059 - 5.0)	0.87 (2.0; 0.028 - 8.3)
2011	2.3 (5.8; 0.038 - 22)	0.10 (0.13; 0.033 - 0.42)	4.6 (8.1; 0.021 - 22)
2012	0.41 (0.92; 0.015 - 3.4)	0.048 (0.016; 0.031 - 0.067)	0.43 (0.95; 0.015 - 3.4)
2013	1.2 (1.0; 0.055 - 1.8)	1.8	0.88 (1.2; 0.055 - 1.7)
BDE-183			
2005	0.62		0.62
2006			
2007	0.74		0.74
2008			
2009	0.10 (0.065; 0.030 - 0.24)	0.24	0.086 (0.052; 0.030 - 0.20)
2010	0.13 (0.11; 0.030 - 0.39)	0.20	0.11 (0.11; 0.030 - 0.39)
2011	0.35 (0.37; 0.088 - 0.61)		0.35 (0.37; 0.088 - 0.61)
2012	0.10 (0.16; 0.014 - 0.43)		0.10 (0.16; 0.014 - 0.43)
2013	0.12		0.12
BDE-209			
2005	1.1 (1.3; 0.11 - 6.5)	1.0 (1.4; 0.15 - 3.2)	0.96 (1.2; 0.11 - 6.5)
2006	1.5 (2.2; 0.084 - 12)	0.85 (0.95; 0.20 - 4.2)	1.0 (2.2; 0.065 - 12)
2007	0.75 (0.68; 0.092 - 3.4)	0.47 (0.71; 0.084 - 3.0)	0.48 (0.40; 0.049 - 1.8)

	Total (GFF+PUF) Mean (STD; Min-Max)	Gas Phase Mean (STD; Min-Max)	Particle Phase Mean (STD; Min-Max)
2008	1.7 (4.6; 0.10 - 25)	0.55 (0.42; 0.15 - 1.2)	1.6 (4.7; 0.087 - 25)
2009	0.81 (0.78; 0.024 - 3.0)	0.26 (0.084; 0.16 - 0.46)	0.72 (0.75; 0.024 - 3.0)
2010	0.50 (0.58; 0.024 - 1.7)		0.50 (0.58; 0.024 - 1.7)
2011	1.2 (0.84; 0.28 - 3.0)		1.2 (0.84; 0.28 - 3.0)
2012	0.34 (0.24; 0.069 - 0.80)		0.34 (0.24; 0.069 - 0.80)
2013	0.12		0.12

Part 2. Egbert from 2005 to 2008

	Total (GFF+PUF) Mean (STD; Min-Max)	Gas Phase Mean (STD; Min-Max)	Particle Phase Mean (STD; Min-Max)
Non-PBDE HFRs			
anti-DDC-CO			
2008	12 (18; 0.40 - 39)		12 (18; 0.40 - 39)
syn-DDC-CO			
2008	6.2 (7.1; 0.47 - 14)		6.2 (7.1; 0.47 - 14)
PBDEs			
BDE-17	0.36 (0.13; 0.55 - 0.16)	0.36 (0.13; 0.16 - 0.55)	
2005	0.38 (0.29; 0.737 - 0.08)	0.34 (0.32; 0.078 - 0.74)	0.50
2006	0.33 (0.21; 0.61 - 0.13)	0.22 (0.13; 0.13 - 0.31)	0.45 (0.24; 0.61 - 0.28)
2007	0.23	0.23	
2008			
BDE-28			
2005	0.88 (1.0; 0.10 - 4.6)	0.92 (1.0; 0.18 - 4.6)	0.096
2006	0.74 (0.74; 0.17 - 2.8)	0.66 (0.67; 0.17 - 2.8)	1.6
2007	0.46 (0.48; 0.12 - 2.0)	0.44 (0.48; 0.12 - 2.0)	0.19
2008	0.27 (0.044; 0.24 - 0.30)	0.27 (0.044; 0.24 - 0.30)	
BDE-47			
2005	9.6 (7.5; 1.2 - 27)	8.5 (7.6; 0.64 - 26)	1.1 (0.7; 0.43 - 3.0)
2006	23 (81; 0.93 - 450)	7.2 (7.7; 0.56 - 37)	16 (80; 0.23 - 440)
2007	3.3 (2.2; 0.73 - 7.8)	2.9 (2.1; 0.66 - 7.5)	0.37 (0.23; 0.072 - 0.94)
2008	3.3 (1.3; 1.7 - 5.8)	1.7 (1.0; 0.85 - 3.5)	1.6 (0.64; 0.60 - 2.3)
BDE-49			
2005	0.48 (0.24; 0.13 - 0.93)	0.50 (0.22; 0.13 - 0.84)	0.11 (0.033; 0.085 - 0.13)
2006	0.91 (1.9; 0.12 - 7.7)	0.44 (0.30; 0.20 - 1.2)	3.7 (5.1; 0.12 - 7.3)
2007	0.33 (0.033; 0.31 - 0.37)	0.33 (0.033; 0.31 - 0.37)	
2008		0.77	
BDE-66			
2005	0.27 (0.15; 0.092 - 0.55)	0.33 (0.13; 0.21 - 0.55)	0.13 (0.050; 0.092 - 0.16)
2006	2.0 (3.4; 0.21 - 8.0)	0.50 (0.24; 0.21 - 0.78)	8.0
2007			
2008			
BDE-71			
2005	0.29		
2006			
2007			
2007			
2008			

	Total (GFF+PUF) Mean (STD; Min-Max)	Gas Phase Mean (STD; Min-Max)	Particle Phase Mean (STD; Min-Max)
BDE-85			
2005			
2006	28		
2007			
2008			
BDE-99			
2005	3.7 (2.1; 0.89 - 10)	2.6 (2.0; 0.57 - 8.6)	1.1 (0.65; 0.33 - 3.6)
2006	22 (100; 0.61 - 560)	2.9 (2.9; 0.43 - 15)	20 (100; 0.21 - 550)
2007	1.6 (0.82; 0.43 - 3.1)	1.1 (0.66; 0.25 - 2.8)	0.46 (0.39; 0.086 - 1.5)
2008	2.6 (0.94; 1.7 - 4.4)	0.94 (0.36; 0.44 - 1.4)	1.6 (0.91; 0.71 - 3.0)
BDE-100			
2005	1.1 (0.70; 0.18 - 2.6)	0.92 (0.68; 0.20 - 2.3)	0.29 (0.16; 0.11 - 0.72)
2006	5.0 (22; 0.13 - 120)	0.89 (0.75; 0.20 - 3.7)	5.6 (25; 0.12 - 120)
2007	0.47 (0.19; 0.087 - 0.71)	0.41 (0.18; 0.12 - 0.69)	0.15 (0.084; 0.052 - 0.27)
2008	0.61 (0.27; 0.20 - 1.0)	0.26 (0.084; 0.16 - 0.36)	0.50 (0.13; 0.30 - 0.66)
BDE-138			
2005	5.1		
2006			
2007			
2008			
BDE-153			
2005	0.60 (0.62; 0.16 - 1.0)		0.60 (0.62; 0.16 - 1.0)
2006	46		46
2007	0.090		0.090
2008			
BDE-154			
2005	0.27 (0.21; 0.14 - 0.51)		0.27 (0.21; 0.14 - 0.51)
2006	8.5 (18; 0.15 - 41)		8.5 (18; 0.15 - 41)
2007			
2008	0.57		0.57
BDE-183			
2005			
2006	0.65		
2007	0.27		
2008			
BDE-209			
2005	9.2 (20; 0.38 - 100)	1.1 (1.7; 0.14 - 5.1)	8.9 (20; 0.23 - 100)
2006	9.7 (14; 0.31 - 52)	4.5 (11; 0.18 - 42)	7.6 (12; 0.31 - 52)
2007	5.6 (6.8; 0.32 - 28)	0.40 (0.038; 0.37 - 0.43)	5.6 (6.8; 0.32 - 28)
2008	34 (46; 4.7 - 130)		34 (46; 4.7 - 130)

281
282

Part 3. Point Petre from 2005 to 2014.

	Total (GFF+PUF) Mean (STD; Min-Max)	Gas Phase Mean (STD; Min-Max)	Particle Phase Mean (STD; Min-Max)
Non-PBDE HFRs			
***anti*-DDC-CO**			
2008	16 (47; 0.085 - 250)	0.093 (0.081; 0.058 - 0.30)	16 (47; 0.085 - 250)
2009	3.0 (7.8; 0.028 - 40)	0.25 (0.40; 0.055 - 1.1)	3.0 (7.8; 0.028 - 40)
2010	2.0 (3.5; 0.058 - 16)	0.06 (0.022; 0.029 - 0.088)	2.0 (3.5; 0.058 - 16)
2011	7.1 (16; 0.14 - 51)		7.1 (16; 0.14 - 51)
2012	6.6 (16; 0.022 - 58)	0.073 (0.055; 0.027 - 0.13)	6.6 (16; 0.022 - 58)
2013	3.1 (5.5; 0.34 - 15)		3.1 (5.5; 0.34 - 15)
2014	1.1 (1.2; 0.28 - 3.3)		1.1 (1.2; 0.28 - 3.3)
***syn*-DDC-CO**			
2008	6.6 (17; 0.082 - 91)	0.10 (0.065; 0.058 - 0.24)	6.6 (17; 0.082 - 91)
2009	1.2 (2.8; 0.030 - 12)	0.081 (0.053; 0.055 - 0.18)	1.1 (2.8; 0.030 - 12)
2010	0.81 (1.3; 0.058 - 5.1)	0.051	0.81 (1.3; 0.057 - 5.1)
2011	4.2 (11; 0.29 - 33)		4.2 (11; 0.29 - 33)
2012	1.6 (3.3; 0.010 - 13)	0.053 (0.036; 0.020 - 0.091)	1.5 (3.3; 0.010 - 13)
2013	0.71 (0.80; 0.21 - 2.1)		0.71 (0.80; 0.21 - 2.1)
2014	0.31 (0.31; 0.14 – 0.74)		0.31 (0.31; 0.14 – 0.77)
TBP-AE			
2009	0.16 (0.11; 0.058 - 0.48)	0.16 (0.11; 0.058 - 0.48)	
2010	1.0 (1.6; 0.089 - 7.0)	1.0 (1.6; 0.089 - 7.0)	0.060 (0.069; 0.029 - 0.27)
2011	1.0 (1.6; 0.042 - 6.2)	1.0 (1.6; 0.042 - 6.2)	0.19
2012	0.41 (0.20; 0.12 - 0.74)	0.40 (0.20; 0.12 - 0.74)	0.025 (0.014; 0.013 - 0.041)
2013	0.15 (0.078; 0.068 - 0.34)	0.15 (0.078; 0.068 - 0.34)	
2014	0.44 (0.44; 0.15 – 1.3)	0.44 (0.44; 0.15 – 1.3)	
TBP-BAE			
2009	0.089 (0.059; 0.059 - 0.18)	0.089 (0.059; 0.059 - 0.18)	0.25
2010	0.094 (0.043; 0.046 - 0.16)	0.090 (0.043; 0.046 - 0.16)	0.057
2011	0.062 (0.031; 0.027 - 0.11)	0.062 (0.031; 0.027 - 0.11)	
2012	0.048 (0.027; 0.024 - 0.12)	0.048 (0.027; 0.024 - 0.12)	0.028
2013	0.31 (0.36; 0.059 - 0.56)	0.31 (0.36; 0.059 - 0.56)	
2014	0.097 (0.021; 0.083 – 0.11)	0.097 (0.021; 0.083 – 0.11)	
BTBPE			
2009			
2010	0.14 (0.12; 0.029 - 0.36)	0.12 (0.019; 0.10 - 0.14)	0.11 (0.12; 0.029 - 0.36)
2011			
2012			
2013			
2014			
DBDPE			
2009			
2010			
2011			

	Total (GFF+PUF) Mean (STD; Min-Max)	Gas Phase Mean (STD; Min-Max)	Particle Phase Mean (STD; Min-Max)
2012	0.31 (0.22; 0.088 - 0.98)		0.31 (0.22; 0.088 - 0.98)
2013	0.73 (0.62; 0.24 - 1.7)		0.73 (0.62; 0.24 - 1.7)
2014			
DPTE			
2009	0.061 (0.00023; 0.060 - 0.061)	0.060 (0.00023; 0.060 - 0.061)	
2010	0.18 (0.17; 0.029 - 0.56)	0.22 (0.18; 0.057 - 0.56)	0.046 (0.021; 0.029 - 0.086)
2011	0.054 (0.025; 0.029 - 0.088)	0.054 (0.025; 0.029 - 0.09)	
2012	0.052 (0.054; 0.018 - 0.17)	0.043 (0.038; 0.018 - 0.13)	0.030 (0.024; 0.013 - 0.047)
2013	0.23 (0.26; 0.050 - 0.76)	0.23 (0.26; 0.050 - 0.76)	
2014	0.50 (0.37; 0.19 – 1.4)	0.50 (0.37; 0.19 – 1.4)	
HBBz			
2009	0.13 (0.17; 0.057 - 0.88)	0.13 (0.17; 0.057 - 0.88)	
2010	0.33 (0.37; 0.029 - 1.9)	0.30 (0.37; 0.045 - 1.8)	0.054 (0.020; 0.029 - 0.12)
2011	0.20 (0.16; 0.049 - 0.55)	0.19 (0.16; 0.049 - 0.55)	0.049 (0.031; 0.023 - 0.083)
2012	0.21 (0.16; 0.028 - 0.57)	0.20 (0.16; 0.028 - 0.57)	0.017 (0.014; 0.0064 - 0.051)
2013	0.25 (0.16; 0.11 - 0.49)	0.25 (0.16; 0.11 - 0.49)	
2014	0.19 (0.11; 0.051 – 0.39)		
HBCDD			
2009	0.16 (0.094; 0.059 - 0.35)		0.16 (0.094; 0.059 - 0.35)
2010	0.31 (0.17; 0.090 - 0.54)		0.31 (0.17; 0.090 - 0.54)
2011	0.38		0.38
2012	0.054 (0.044; 0.023 - 0.085)		0.054 (0.044; 0.023 - 0.085)
2013			
2014			
PBEB			
2009	0.060 (0.032; 0.024 - 0.14)	0.060 (0.032; 0.024 - 0.14)	
2010	0.045 (0.030; 0.014 - 0.12)	0.045 (0.030; 0.014 - 0.12)	
2011	0.034 (0.023; 0.012 - 0.087)	0.034 (0.023; 0.012 - 0.087)	
2012	0.11 (0.067; 0.063 - 0.16)	0.11 (0.067; 0.063 - 0.16)	
2013			
2014	0.022	0.022	
PBT			
2009		0.059 (0.0010; 0.058 - 0.061)	
2010		0.071 (0.047; 0.013 - 0.18)	
2011		0.050 (0.032; 0.0080 - 0.12)	
2012		0.050 (0.027; 0.014 - 0.10)	
2013		0.22 (0.16; 0.036 - 0.52)	
2014		0.083 (0.046; 0.034 – 0.17)	
EH-TBB			
2009	0.60 (0.10; 0.53 - 0.67)		0.60 (0.10; 0.53 - 0.67)
2010	6.0 (5.4; 0.15 - 14)	7.0 (5.2; 0.80 - 14)	0.13
2011	2.5 (2.3; 0.98 - 5.9)	3.0 (2.6; 0.98 - 5.9)	1.3

	Total (GFF+PUF) Mean (STD; Min-Max)	Gas Phase Mean (STD; Min-Max)	Particle Phase Mean (STD; Min-Max)
2012	0.46 (0.36; 0.051 - 1.2)	0.38 (0.23; 0.075 - 0.78)	0.24 (0.39; 0.051 - 1.2)
2013			
2014	2.4 (2.1; 0.32 – 4.5)	3.4 (1.5; 2.4 – 4.5)	0.32
PBDEs			
BDE-17			
2005	0.42 (0.17; 0.14 - 0.70)	0.42 (0.17; 0.14 - 0.70)	
2006	0.36 (0.19; 0.16 - 0.71)	0.36 (0.19; 0.16 - 0.71)	
2007			
2008	0.18 (0.14; 0.057 - 0.57)	0.17 (0.14; 0.057 - 0.57)	0.088 (0.0002; 0.088 - 0.088
2009	0.11 (0.062; 0.030 - 0.24)	0.11 (0.058; 0.056 - 0.24)	0.030 (0.00019; 0.029 - 0.030)
2010	0.14 (0.10; 0.056 - 0.49)	0.14 (0.10; 0.056 - 0.49)	0.029 (0.00068; 0.029 - 0.030)
2011	0.12 (0.062; 0.043 - 0.21)	0.12 (0.062; 0.043 - 0.21)	
2012	0.10 (0.074; 0.018 - 0.26)	0.10 (0.072; 0.018 - 0.25)	0.010 (0.0048; 0.0068 - 0.014)
2013	0.19 (0.080; 0.10 - 0.28)	0.19 (0.080; 0.10 - 0.28)	
2014	0.14 (0.089; 0.015 – 0.26)	0.17 (0.068; 0.10 – 0.26)	0.015
BDE-28			
2005	0.74 (0.39; 0.24 - 1.6)	0.74 (0.39; 0.24 - 1.6)	
2006	0.63 (0.30; 0.29 - 1.4)	0.63 (0.30; 0.29 - 1.4)	
2007	0.36 (0.11; 0.21 - 0.54)	0.38 (0.11; 0.24 - 0.54)	0.21
2008	0.31 (0.27; 0.058 - 1.0)	0.29 (0.27; 0.057 - 1.0)	0.086 (0.0040; 0.080 - 0.091)
2009	0.22 (0.16; 0.055 - 0.6)	0.21 (0.16; 0.055 - 0.6)	0.030 (0.00071; 0.028 - 0.030)
2010	0.25 (0.26; 0.046 - 1.2)	0.24 (0.26; 0.046 - 1.2)	0.034 (0.011; 0.029 - 0.057)
2011	0.24 (0.21; 0.022 - 0.63)	0.24 (0.21; 0.020 - 0.63)	
2012	0.32 (0.27; 0.045 - 0.89)	0.31 (0.27; 0.041 - 0.88)	0.0062 (0.0048; 0.0019 - 0.021)
2013	0.34 (0.29; 0.065 - 1.0)	0.34 (0.29; 0.065 - 0.98)	
2014	0.40 (0.29; 0.024 – 0.87)	0.47 (0.25; 0.23 – 0.87)	0.024
BDE-47			
2005	9.8 (7.1; 2.6 - 28)	8.7 (7.2; 1.6 - 27)	1.1 (0.88; 0.31 - 3.9)
2006	6.6 (5.4; 0.49 - 24)	6.2 (5.4; 0.75 - 23)	0.63 (0.44; 0.22 - 2.6)
2007	5.0 (7.7; 1.5 - 43)	3.2 (3.3; 0.95 - 17)	1.9 (7.1; 0.27 - 39)
2008	5.8 (4.0; 1.9 - 17)	4.1 (3.3; 1.3 - 12)	1.8 (3.0; 0.41 - 15)
2009	3.5 (2.3; 0.98 - 10)	2.9 (2.3; 0.80 - 9.2)	0.55 (0.51; 0.062 - 2.4)
2010	2.7 (1.9; 0.20 - 9.2)	2.4 (1.9; 0.50 - 9.0)	0.33 (0.43; 0.090 - 2.5)
2011	2.7 (1.1; 0.99 - 4.7)	2.4 (1.1; 0.80 - 4.5)	0.23 (0.11; 0.11 - 0.52)
2012	2.9 (1.4; 0.99 - 5.6)	2.7 (1.3; 0.84 - 5.5)	0.23 (0.30; 0.071 - 1.2)
2013	2.5 (1.3; 1.0 - 5.1)	2.3 (1.3; 0.86 - 4.9)	0.27 (0.22; 0.091 - 0.81)
2014	4.6 (1.9; 1.8 – 8.7)	4.2 (4.8; 1.5 – 8.0)	0.43 (0.16; 0.29 – 0.77)
BDE-49			
2005	0.49 (0.29; 0.11 - 1.2)	0.57 (0.26; 0.29 - 1.2)	0.11 (0.037; 0.062 - 0.15)
2006	0.40 (0.18; 0.14 - 0.72)	0.40 (0.18; 0.14 - 0.72)	
2007	0.39 (0.34; 0.12 - 1.1)	0.25 (0.10; 0.12 - 0.36)	1.1
2008	0.22 (0.13; 0.058 - 0.54)	0.17 (0.14; 0.057 - 0.54)	0.11 (0.071; 0.080 - 0.34)
2009	0.14 (0.090; 0.058 - 0.40)	0.12 (0.087; 0.055 - 0.36)	0.035 (0.012; 0.028 - 0.060)
2010	0.13 (0.12; 0.029 - 0.58)	0.13 (0.12; 0.051 - 0.58)	0.044 (0.045; 0.029 - 0.17)
2011	0.15 (0.075; 0.066 - 0.26)	0.15 (0.075; 0.066 - 0.26)	

	Total (GFF+PUF) Mean (STD; Min-Max)	Gas Phase Mean (STD; Min-Max)	Particle Phase Mean (STD; Min-Max)
2012	0.14 (0.091; 0.028 - 0.30)	0.13 (0.085; 0.022 - 0.28)	0.013 (0.021; 0.0032 - 0.081)
2013	0.16 (0.11; 0.10 - 0.35)	0.16 (0.11; 0.10 - 0.35)	
2014	0.18 (0.098; 0.027 – 0.28	0.21 (0.071; 0.12 – 0.28)	0.027
BDE-66			
2005	0.13		0.13
2006			
2007	1.1		1.1
2008	0.18 (0.13; 0.057 - 0.59)	0.15 (0.13; 0.057 - 0.59)	0.12 (0.088; 0.080 - 0.34)
2009	0.10 (0.069; 0.055 - 0.35)	0.080 (0.042; 0.055 - 0.18)	0.073 (0.089; 0.029 - 0.29)
2010	0.14 (0.16; 0.029 - 0.59)	0.15 (0.18; 0.046 - 0.59)	0.058 (0.064; 0.029 - 0.17)
2011	0.10 (0.030; 0.072 - 0.13)	0.10 (0.030; 0.072 - 0.13)	
2012	0.075 (0.063; 0.0056 - 0.22)	0.084 (0.048; 0.023 - 0.15)	0.019 (0.024; 0.0048 - 0.071)
2013			
2014			
BDE-71			
2005	0.29		0.29
2006			
2007			
2008	0.22 (0.18; 0.088 - 0.43)	0.059	0.20 (0.20; 0.088 - 0.43)
2009	0.058	0.058	
2010			
2011			
2012	0.067	0.067	
2013			
2014			
BDE-85			
2005	0.12		0.12
2006			
2007	1.9 (2.2; 0.35 - 3.4)	0.35	
2008	0.28 (0.50; 0.057 - 1.7)	0.16 (0.32; 0.057 - 1.5)	0.35 (0.59; 0.084 - 1.6)
2009	0.069 (0.040; 0.029 - 0.20)	0.072 (0.034; 0.055 - 0.18)	0.042 (0.016; 0.029 - 0.060)
2010	0.18 (0.37; 0.029 - 1.8)	0.21 (0.43; 0.055 - 1.8)	0.080 (0.064; 0.029 - 0.23)
2011	0.067	0.067	
2012	0.069 (0.054; 0.016 - 0.18)	0.053 (0.032; 0.020 - 0.10)	0.049 (0.066; 0.0078 - 0.18)
2013			
2014	0.74 (0.21; 0.55 – 0.97)	0.74 (0.21; 0.55 – 0.97)	
BDE-99			
2005	4.2 (2.0; 1.6 - 8.3)	3.2 (2.1; 0.76 - 7.5)	1.1 (0.85; 0.30 - 3.4)
2006	3.0 (1.8; 0.69 - 8.6)	2.2 (1.5; 0.31 - 7.2)	0.78 (1.0; 0.092 - 5.2)
2007	4.3 (12; 0.77 - 69)	1.5 (2.4; 0.34 - 11)	3.0 (13; 0.24 - 68)
2008	5.0 (8.0; 1.2 - 37)	2.1 (2.9; 0.70 - 17)	3.1 (7.7; 0.33 - 36)
2009	1.5 (0.48; 0.82 - 2.7)	1.1 (0.46; 0.52 - 2.6)	0.40 (0.31; 0.040 - 1.3)
2010	2.0 (1.2; 0.38 - 7.1)	1.6 (0.75; 0.55 - 3.9)	0.46 (0.94; 0.090 - 5.2)
2011	1.8 (0.40; 1.3 - 2.5)	1.6 (0.40; 1.12 - 2.3)	0.25 (0.17; 0.090 - 0.78)

	Total (GFF+PUF) Mean (STD; Min-Max)	Gas Phase Mean (STD; Min-Max)	Particle Phase Mean (STD; Min-Max)
2012	2.1 (1.7; 0.66 - 6.1)	1.5 (0.81; 0.57 - 3.2)	0.61 (1.2; 0.059 - 4.6)
2013	1.3 (0.43; 0.32 - 1.9)	1.1 (0.46; 0.23 - 1.8)	0.27 (0.27; 0.062 - 0.79)
2014	3.4 (3.8; 0.58 – 12)	3.2 (3.7; 0.46 – 12)	0.17 (0.075; 0.090 – 0.34)
BDE-100			
2005	1.0 (0.54; 0.34 - 2.3)	0.91 (0.59; 0.24 - 2.1)	0.30 (0.20; 0.077 - 0.80)
2006	0.78 (0.56; 0.070 - 2.6)	0.79 (0.54; 0.19 - 2.6)	0.21 (0.16; 0.070 - 0.73)
2007	1.1 (2.8; 0.085 - 14)	0.66 (0.66; 0.17 - 2.5)	1.5 (4.1; 0.085 - 13)
2008	1.2 (1.6; 0.35 - 7.2)	0.6 (0.88; 0.23 - 5.1)	0.63 (1.4; 0.080 - 6.9)
2009	0.44 (0.19; 0.17 - 0.97)	0.33 (0.17; 0.12 - 0.85)	0.11 (0.085; 0.029 - 0.35)
2010	0.46 (0.24; 0.087 - 1.4)	0.37 (0.16; 0.13 - 0.81)	0.10 (0.16; 0.029 - 0.92)
2011	0.41 (0.086; 0.26 - 0.56)	0.37 (0.081; 0.21 - 0.49)	0.064 (0.034; 0.031 - 0.14)
2012	0.46 (0.27; 0.20 - 1.0)	0.36 (0.15; 0.17 - 0.69)	0.10 (0.18; 0.015 - 0.65)
2013	0.26 (0.12; 0.14 - 0.49)	0.27 (0.13; 0.15 - 0.49)	0.11 (0.065; 0.047 - 0.19)
2014	0.76 (0.75; 0.13 – 2.4)	0.73 (0.72; 0.13 – 2.3)	0.066 (0.017; 0.040 – 0.081)
BDE-138			
2005			
2006			
2007			
2008	0.26 (0.19; 0.059 - 0.43)	0.18 (0.20; 0.059 - 0.41)	0.39 (0.062; 0.34 - 0.43)
2009	0.075 (0.057; 0.029 - 0.18)	0.10 (0.069; 0.055 - 0.18)	0.044 (0.022; 0.029 - 0.060)
2010			0.057
2011			
2012	0.035 (0.013; 0.020 - 0.047)		0.035 (0.013; 0.020 - 0.047)
2013			
2014	0.42	0.42	
BDE-153			
2005	0.44 (0.23; 0.30 - 0.71)		0.44 (0.23; 0.30 - 0.71)
2006			
2007	3.9 (4.4; 0.75 – 7.0)		7.0
2008	0.39 (0.70; 0.057 - 3.1)	0.20 (0.46; 0.057 - 2.2)	0.38 (0.72; 0.084 - 3.0)
2009	0.088 (0.046; 0.029 - 0.23)	0.065 (0.028; 0.055 - 0.18)	0.056 (0.034; 0.028 - 0.15)
2010	0.13 (0.14; 0.029 - 0.74)	0.080 (0.031; 0.043 - 0.15)	0.094 (0.14; 0.029 - 0.63)
2011	0.086	0.086	
2012	0.11 (0.13; 0.010 - 0.44)	0.067 (0.034; 0.030 - 0.13)	0.085 (0.14; 0.010 - 0.44)
2013	0.084 (0.0099; 0.077 - 0.091)		0.084 (0.010; 0.077 - 0.091)
2014	1.6 (0.95; 0.39 – 2.6)	1.6 (0.95; 0.39 – 2.6)	
BDE-154			
2005	0.38 (0.10; 0.28 - 0.48)		0.38 (0.10; 0.28 - 0.48)
2006	0.39 (0.18; 0.22 - 0.58)		0.39 (0.18; 0.22 - 0.58)
2007	3.5 (3.9; 0.75 - 6.3)		6.4
2008	0.43 (0.79; 0.057 - 2.9)	0.18 (0.46; 0.057 - 2.5)	0.40 (0.81; 0.084 - 2.8)
2009	0.095 (0.044; 0.029 - 0.23)	0.065 (0.024; 0.055 - 0.18)	0.048 (0.026; 0.028 - 0.12)
2010	0.12 (0.080; 0.030 - 0.46)	0.091 (0.035; 0.033 - 0.18)	0.068 (0.081; 0.029 - 0.34)
2011	0.076 (0.042; 0.045 - 0.15)	0.080 (0.048; 0.045 - 0.15)	0.063

	Total (GFF+PUF) Mean (STD; Min-Max)	Gas Phase Mean (STD; Min-Max)	Particle Phase Mean (STD; Min-Max)
2012	0.12 (0.14; 0.0054 - 0.48)	0.078 (0.048; 0.030 - 0.18)	0.059 (0.12; 0.0035 - 0.43)
2013	0.064		0.064
2014	1.0 (0.79; 0.24 – 2.0)	1.0 (0.79; 0.24 – 2.0)	
BDE-183			
2005	1.9		1.9
2006			
2007			
2008	0.41 (0.55; 0.059 - 2.0)	0.78 (0.76; 0.059 - 2.0)	0.17 (0.15; 0.084 - 0.52)
2009	0.21 (0.37; 0.029 - 1.2)	0.82 (0.56; 0.18 - 1.22)	0.063 (0.034; 0.029 - 0.12)
2010	0.089 (0.043; 0.029 - 0.15)		0.089 (0.043; 0.029 - 0.15)
2011	0.41		0.41
2012	0.063 (0.028; 0.027 - 0.095)		0.063 (0.028; 0.027 - 0.095)
2013			
2014			
BDE-209			
2005	6.1 (17; 0.30 - 72)	1.6 (1.5; 0.43 - 3.4)	5.8 (17; 0.30 - 72)
2006	1.3 (0.97; 0.14 - 3.4)	0.52 (0.39; 0.10 - 1.4)	1.0 (0.77; 0.14 - 3.0)
2007	1.7 (2.2; 0.18 - 12)	0.27 (0.22; 0.078 - 0.95)	1.6 (2.2; 0.18 - 12)
2008	5.2 (3.4; 0.68 - 14)	0.35 (0.16; 0.24 - 0.46)	5.2 (3.4; 0.68 - 14)
2009	1.6 (1.6; 0.072 - 7.5)	0.18 (0.044; 0.11 - 0.24)	1.6 (1.6; 0.072 - 7.5)
2010	0.68 (0.74; 0.017 - 3.2)		0.68 (0.74; 0.017 - 3.2)
2011	1.7 (1.1; 0.46 - 4.6)		1.7 (1.1; 0.46 - 4.6)
2012	0.90 (1.5; 0.055 - 4.5)		0.90 (1.5; 0.055 - 4.5)
2013	1.4 (0.78; 0.57 - 2.6)		1.4 (0.78; 0.57 - 2.6)
2014	1.8 (0.32; 1.5 – 2.2)		1.8 (0.32; 1.5 – 2.2)

Table S4. Pearson correlation coefficients among the primary components of the Penta BDE formulations at Point Petre, Burnt Island and Egbert. The primary components of the PentaBDE technical mixtures are significantly ($p < 0.05$) correlated.

Point Petre	**BDE-47**	**BDE-99**	**BDE-100**
BDE-47	1.0		
BDE-99	0.68	1.0	
BDE-100	0.75	0.98	1.0
Burnt Island			
BDE-47	1.0		
BDE-99	0.96	1.0	
BDE-100	0.96	0.99	1.0
Egbert			
BDE-47	1.0		
BDE-99	0.99	1.0	
BDE-100	0.99	0.99	1.0

Table S5. Ratios between BDE-47 and -99, BDE-99 and -100 and anti-DDC-CO and total DDC-CO in Great Lakes air

	Burnt Island	Egbert	Point Petre
BDE-47/BD-E99			
2005	2.2	2.4	2.2
2006	1.7	1.9	2.2
2007	2.7	2.2	2.0
2008	2.8	1.3	1.7
2009	2.1		2.2
2010	1.6		1.4
2011	2.4		1.5
2012	3.0		1.9
2013	0.9		2.0
2014			3.3
BDE-99/BDE-100			
2005	4.6	4.1	4.2
2006	4.5	4.4	5.2
2007	5.5	4.1	6.1
2008	4.4	4.7	3.8
2009	4.0		3.6
2010	4.4		4.2
2011	4.0		4.4
2012	3.5		4.3
2013	4.7		6.0
2014			4.1
***anti*-DDC-CO/Total DDC-CO**			
2008	0.72		0.67
2009	0.72		0.76
2010	0.76		0.79
2011	0.68		0.79
2012	0.80		0.68
2013	0.79		0.84
2014			0.74

312 **Table S6.** Regression results from the one-parameter non-linear model

Sampling period	A x 10^5	R^2
04-02-2005	0.42	0.40
17-04-2005	1.2	0.98
11-05-2005	0.94	0.99
04-06-2005	0. 25	0.99
28-06-2005	0.00015	0.31
10-07-2005	0. 22	0.99
22-07-2005	0. 79	0.98
03-08-2005	0. 33	0.98
15-08-2005	0. 097	0.99
27-08-2005	0. 087	0.99
08-09-2005	0. 10	0.99
20-09-2005	0. 15	0.99
02-10-2005	0. 15	0.99
14-10-2005	0. 049	0.99
25-12-2005	0.79	0.03
30-05-2005	0.054	0.23
23-06-2006	0. 069	0.99
17-07-2006	0. 29	0.95
29-07-2006	0. 13	0.33
10-08-2006	0. 093	0.99
15-09-2006	0. 13	0.99
09-10-2006	0. 20	0.95
24-07-2007	0. 13	0.89
05-08-2007	0. 0016	0.99
22-09-2007	0. 34	0.97
13-02-2008	0.0016	0.19
25-02-2008	0. 23	0.55
08-03-2008	0.20	0.58
20-03-2008	3.5	0.89
01-04-2008	3.6	0.87
25-04-2008	0.25	0.75
07-05-2008	0.53	0.90
19-05-2008	0.0012	0.91
31-05-2008	1.7	0.87
24-06-2008	0.67	0.90
06-07-2008	1.1	0.90
18-07-2008	1.4	0.75
30-07-2008	0.019	0.46
12-08-2008	0.019	0.46

Sampling period	A x 10^5	R^2
23-08-2008	0.65	0.97
04-09-2008	0.012	0.76
28-09-2008	0.0085	0.96
22-10-2008	0.0012	0.83
03-11-2008	0.80	0.72
15-11-2008	0.81	0.86
27-11-2008	0.18	0.77
21-12-2008	0.044	0.74
07-02-2009	0.073	0.21
19-02-2009	0.23	0.68
15-03-2009	0.36	0.40
08-04-2009	0.0036	0.22
02-05-2009	0.015	0.87
14-05-2009	0.039	0.93
26-05-2009	0.00016	0.94
07-06-2009	0.27	0.79
19-06-2009	0.084	0.89
25-07-2009	0.40	0.86
06-08-2009	0.15	0.78
18-08-2009	0.30	0.95
30-08-2009	0.026	0.90
11-09-2009	0.50	0.63
23-09-2009	1.6	0.86
05-10-2009	0.50	0.78
17-10-2009	0.13	0.71
29-10-2009	0.22	0.62
10-11-2009	0.25	0.74
22-11-2009	0.074	0.18
04-12-2009	0.023	0.97
16-12-2009	0.0073	0.85
28-12-2009	0.17	0.82
09-01-2010	0.19	0.40
21-01-2010	0.018	0.10
02-02-2010	0.00030	0.85
14-02-2010	0.0023	0.73
26-02-2010	0.00032	0.92
10-03-2010	0.12	0.69
03-04-2010	0.31	0.92
15-04-2010	0.031	0.77
27-04-2010	0.0014	0.96

Sampling period	A x 10^5	R^2
09-05-2010	0.0033	0.99
02-06-2010	2.9	0.23
14-06-2010	0.35	0.56
26-06-2010	0.43	0.38
08-07-2010	0.23	0.81
20-07-2010	0.12	0.87
13-08-2010	0.098	0.90
06-09-2010	0.16	0.96
18-09-2010	0.24	0.57
24-10-2010	0.0099	0.91
05-11-2010	0.061	0.62
17-11-2010	0.051	0.95
16-01-2011	0.033	0.93
05-03-2011	0.071	0.98
09-06-2011	0.36	0.99
03-07-2011	0.040	0.99
20-08-2011	0.67	0.99
13-09-2011	0.45	0.99
07-10-2011	0.038	0.17
31-10-2011	0.0038	0.93
18-12-2011	0.00016	0.84
11-01-2012	0.33	0.95
04-02-2012	0.089	0.30
28-02-2012	0.013	0.02
23-02-2012	0.012	0.98
16-04-2012	0.028	0.89
10-05-2012	0.22	0.87
03-06-2012	0.56	0.90
27-06-2012	1.2	0.57
14-08-2012	0.13	0.99
07-09-2012	0.63	0.85
01-10-2012	0.0055	0.99
25-10-2012	0.057	0.94
18-11-2012	0.051	0.82
12-12-2012	0.0027	0.02

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Table S7. First order apparent half-lives ($t_{1/2}$) of halogenated flame retardants at the three Canadian Great Lakes sites.

	Burnt Island (2005-2013)		Burnt Island (2005-2013)		Point Petre (2005-2014)		Point Petre (2005-2014)	
BDEs	**$t_{1/2}$ (year)**	**r^2**	**t_2 (year)**	**r^2**	**$t_{1/2}$ (year)**	**r^2**	**t_2 (year)**	**r^2**
BDE-17	8.1	0.75			8.1	0.43		
BDE-28	26	0.36			15	0.28		
BDE-47			23	0.39	7.2	0.62		
BDE-49	8.4	0.47			5.9	0.72		
BDE-99	13	0.44			6.6	0.85		
BDE-100			17	0.93	5.9	0.94		
BDE-153*	2.5	0.62					5.0	0.24
BDE-154*	1.8	0.81				0.95	5.7	0.24
BDE-209	5.7	0.96			16	0.12		
Total BDE			20	0.53	8.4	0.79		
non-PBDE HFRs	**(2008-2013)**		**(2008-2013)**		**(2008-2014)**		**(2008-2014)**	
anti-DDC-CO	3.4	0.36			13	0.11		
syn-DDC-CO			7.6	0.21	6.6	0.41		
TBP-AE	24	0.1					14	0.1
HBBz	27	0.11					11	0.33
PBT	2.1	0.99					7.6	0.55

*Half-life was derived for BDE-153 and BDE-154 at Burnt Island from 2008 to 2013 and BDE-153 and BDE-154 at Point Petre from 2008 to 2014

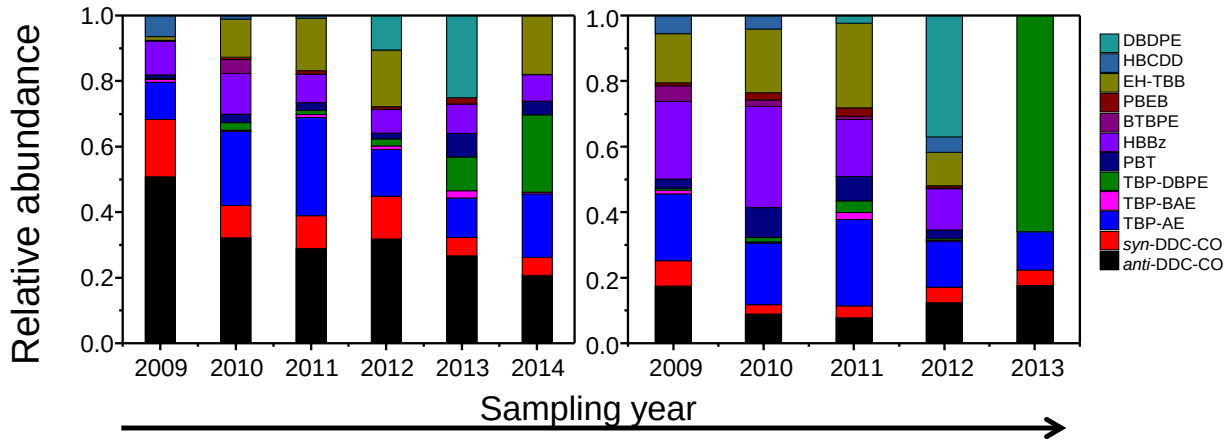

Figure S1. Relative abundance of non-PBDE HFRs at Point Petre and Burnt Island.

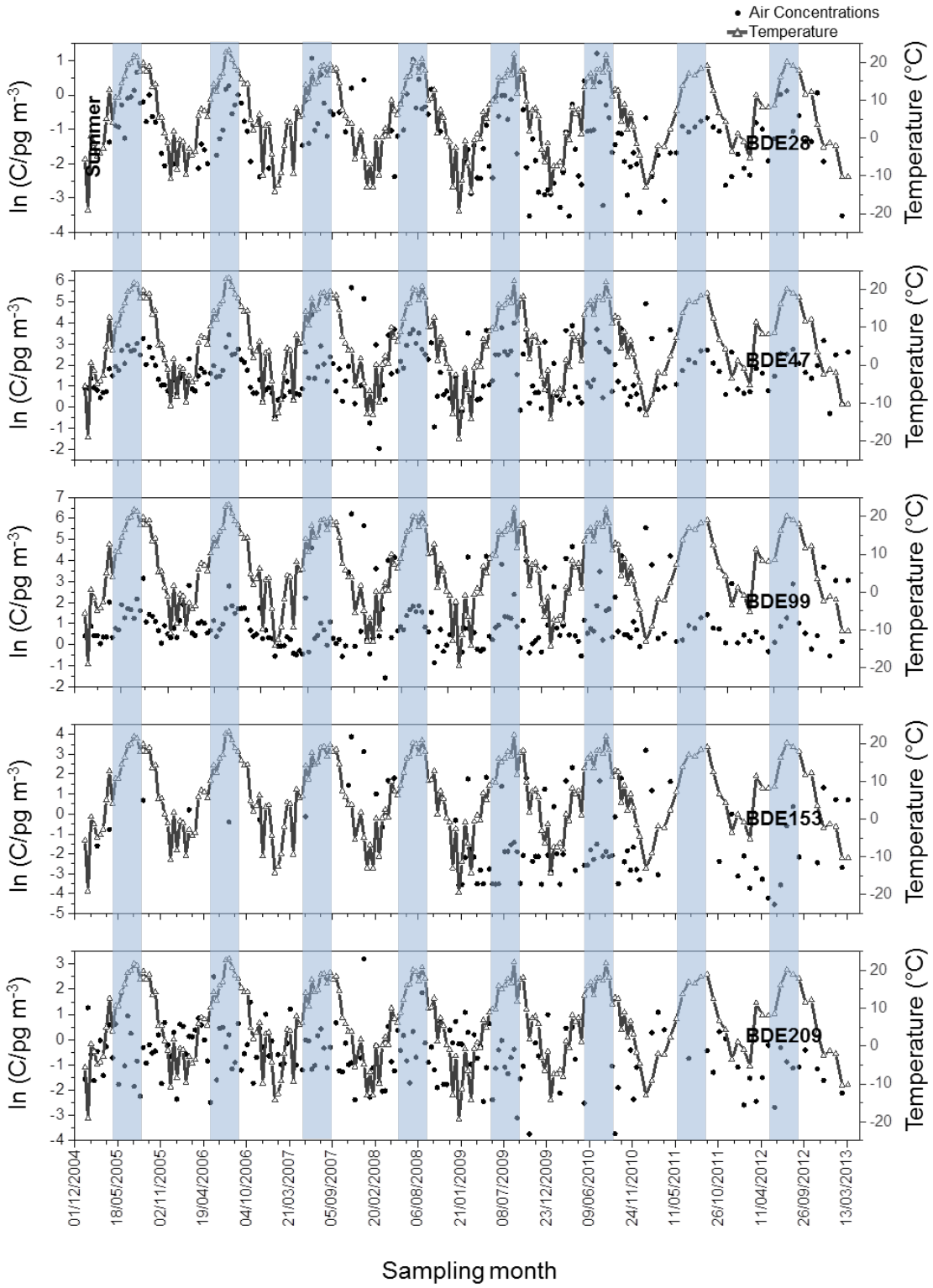

Figure S2. Seasonal profiles of air concentrations of PBDEs and ambient temperature at Burnt Island. Blue shadings indicate summer period (April to September).

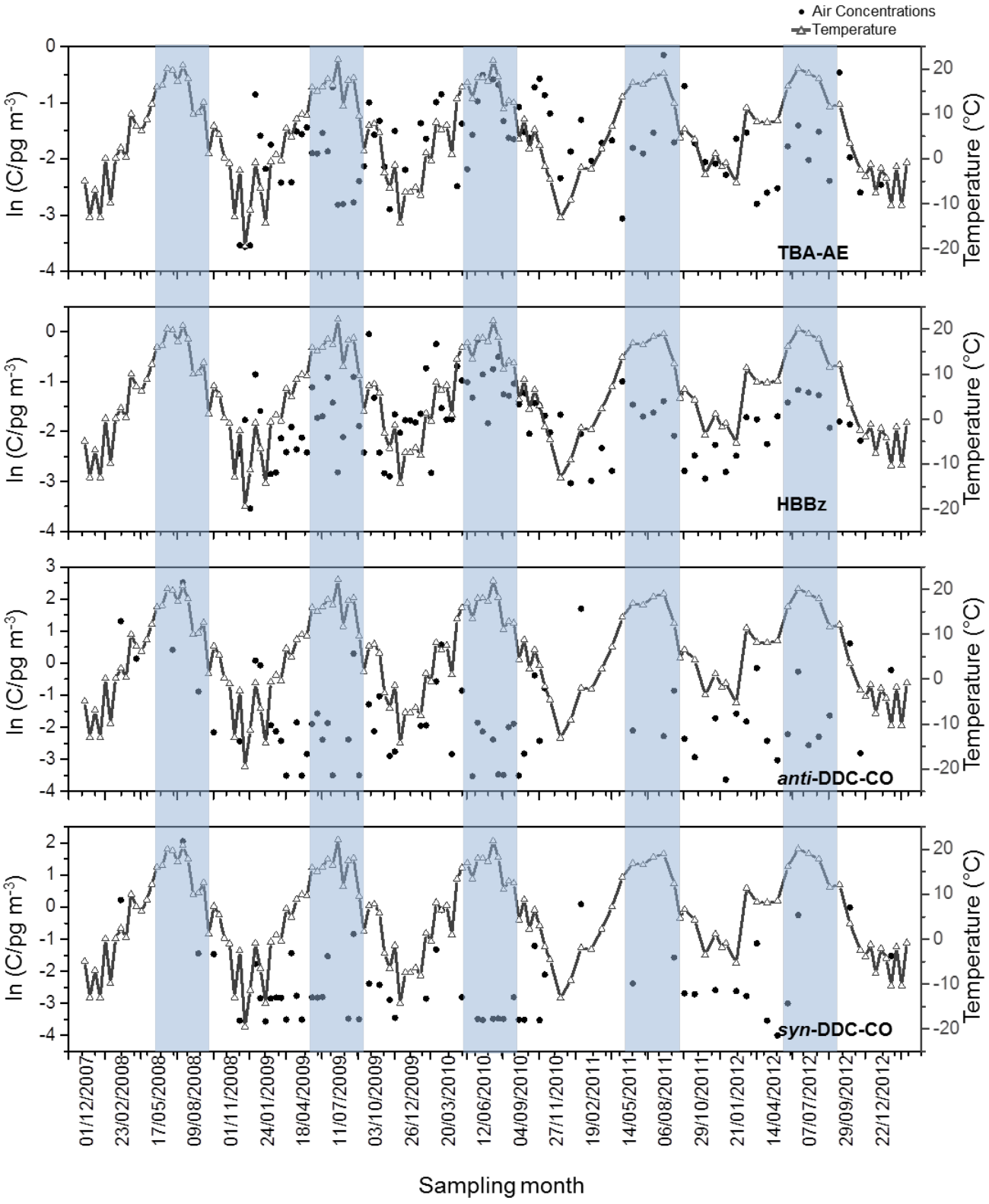

Figure S3. Seasonal profiles of air concentrations of non-PBDE HFRs and ambient temperature at Burnt Island. Blue shadings indicate summer period (April to September).

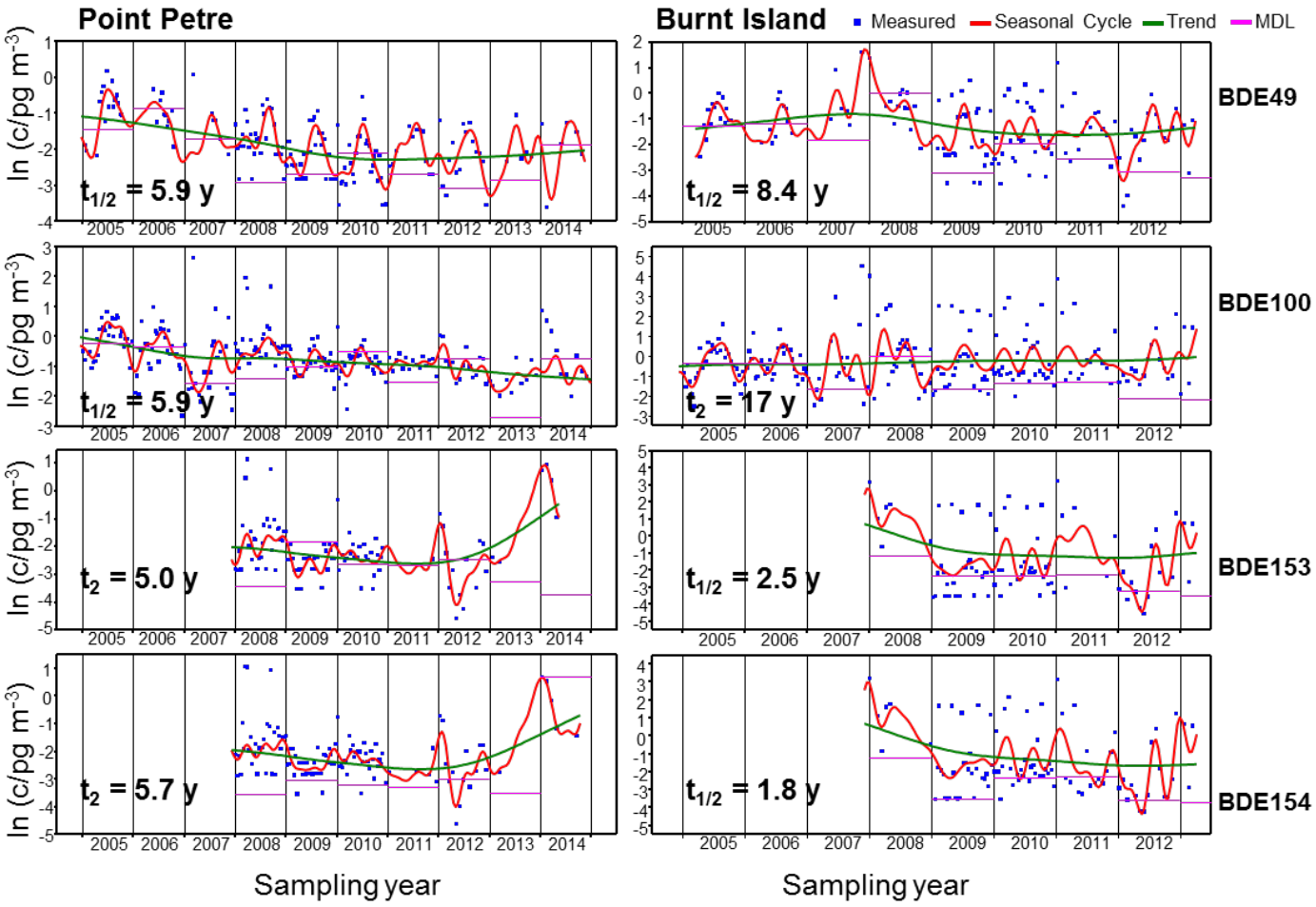

Figure S4. Seasonal cycles, trends and measurements of BDE-49, -100, -153 and -154 at Burnt Island and Point Petre.

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