A Retrospective Analysis of Mussel Monitoring for Nearshore Toxics in Puget Sound

Draft Report

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1 Background

This report provides a retrospective analysis of data generated by previous mussel monitoring surveys coordinated under Washington Department of Fish and Wildlife's (WDFW) Toxics Biological Obsevation System (TBiOS). We determine how existing contaminant data from historical bay mussels (*Mytilus trossulus*) can be used as a Toxics in the Nearshore Vital Sign indicator. In addition, we assess the predictive ability of existing sampling rate to predict expected contaminant trends.

2 Methods

All materials used to prepare this report can be found in the following GitHub repository: `https://github.com/cwangen/mussel_toxics`.

2.1 Data

Uncontaminated mussels from a local aquaculture source were transplanted to various locations along the Puget Sound shoreline, covering a broad range of upland land-use types from rural to highly urban. Mussels were placed into cages and left to incubate for 49-107 days before they were recovered, and concentrations of several major contaminant classes were measured. Four mussels surveys were performed, with mussels being retrieved in 2013, 2016, 2018, and 2020. Our analysis focuses on polycyclic aromatic hydrocarbons (PAHs), polybrominated diphenyl ethers (PBDEs), and polychlorinated biphenyls(PCBs) due to their significance in both ecosystem and human health.

2.2 Data Analysis

The data used in this analysis originated in the form of an Microsoft Excel file, titled `2013-20MusselCagesPOPsPAHs_Cnty_WRIA_LIO_Coverages.xlsx`. The data included more fields than used in our analysis, and can be found in its entirety in `/mussel_toxics/data/raw/`. The data file was cleaned in order to correct minor inconsistencies and errors, resulting in `/mussel_toxics/data/clean/totals_all.csv`.

We used the dry weights of toxics found in each sample for analysis. Although the data records also included the concentration of lipids, as well as wet weight, dry weight is the standard for reporting toxicology data. Data summaries include the following

- Tables of dry weights organized by categorical variables;
- Maps of dry weight concentrations;
- Raincloud plots, consisting of a box plot, an approximate probability density, and the individual data points, for each toxin for based upon WRIA and year.

Samples from the original aquaculture source used as reference materials were removed from the analyses, but included in the concentration maps. Samples outside of Puget Sound (east of -123.5 longitude) were also removed from sampling, but remain in map figures. Figures 1, 2, and 3 display the concentrations of PAHs, PBDEs, and PCBs over time in mussel samples, respectively. They also shed light on how sampling is inconsistent both spatially and temporally.

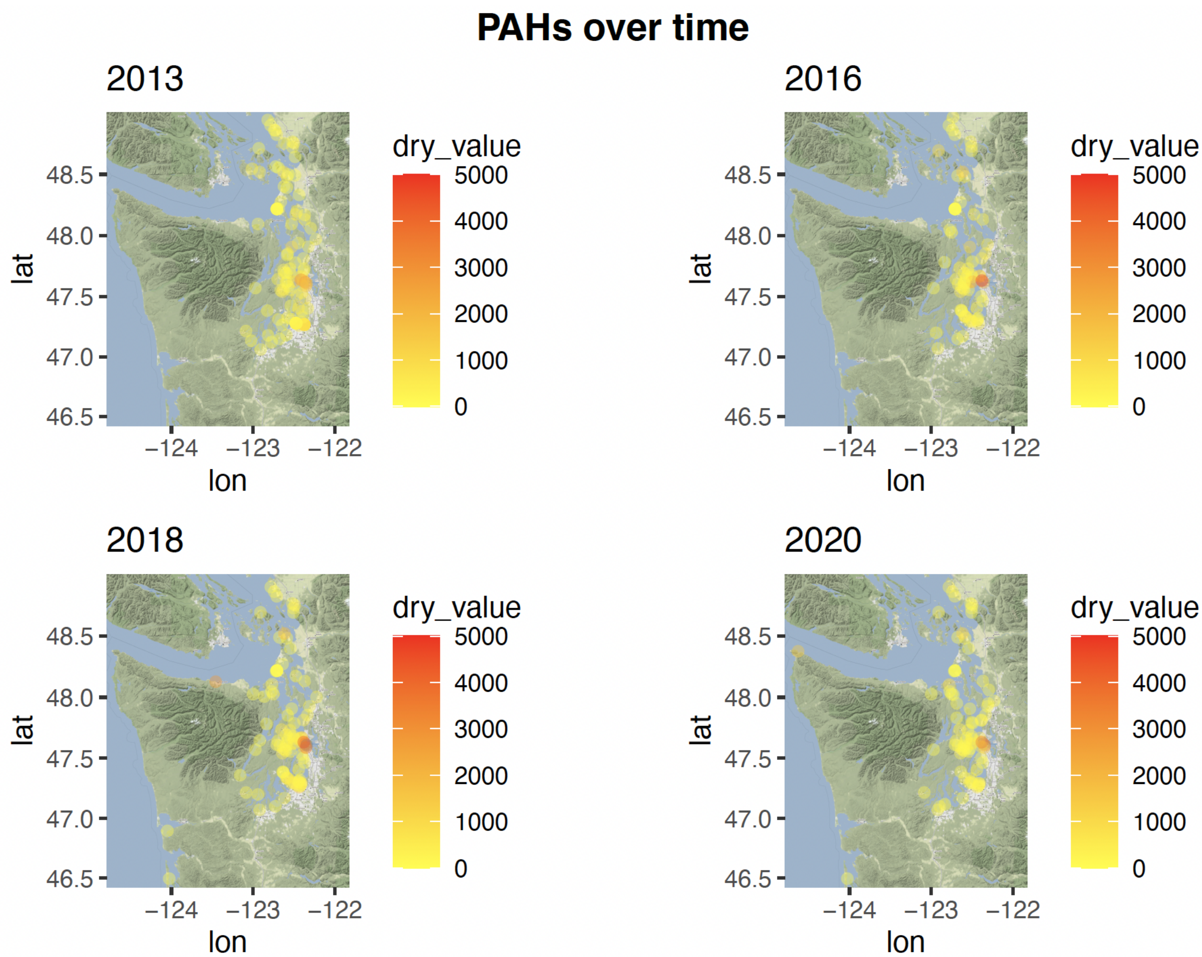


Figure 1: Estimated concentration of PAHs across Puget Sound.

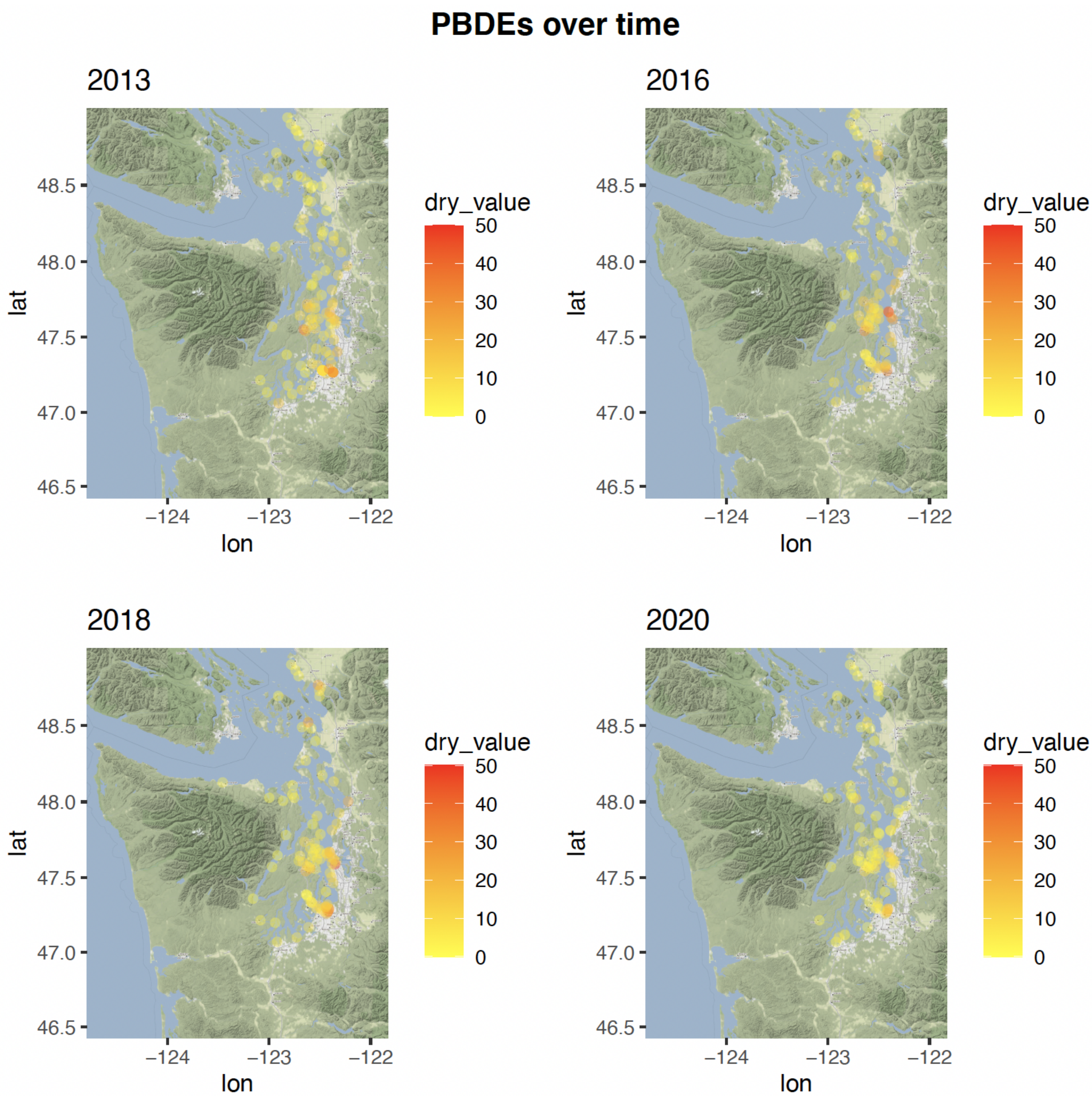


Figure 2: Estimated concentration of PBDEs across Puget Sound.

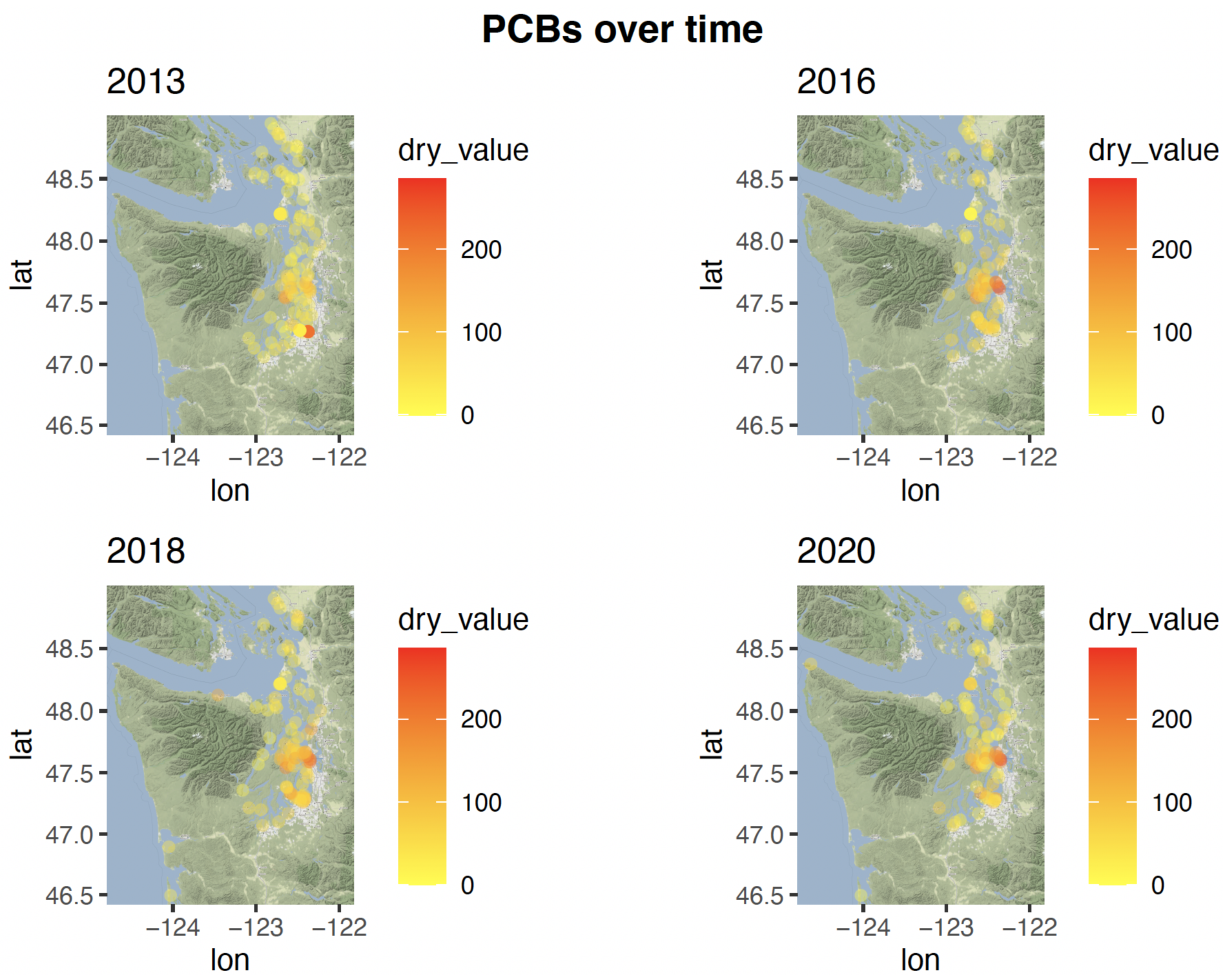


Figure 3: Estimated concentration of PCBs across Puget Sound.

Figure 4: Distribution of the log concentration of PAHs across years.

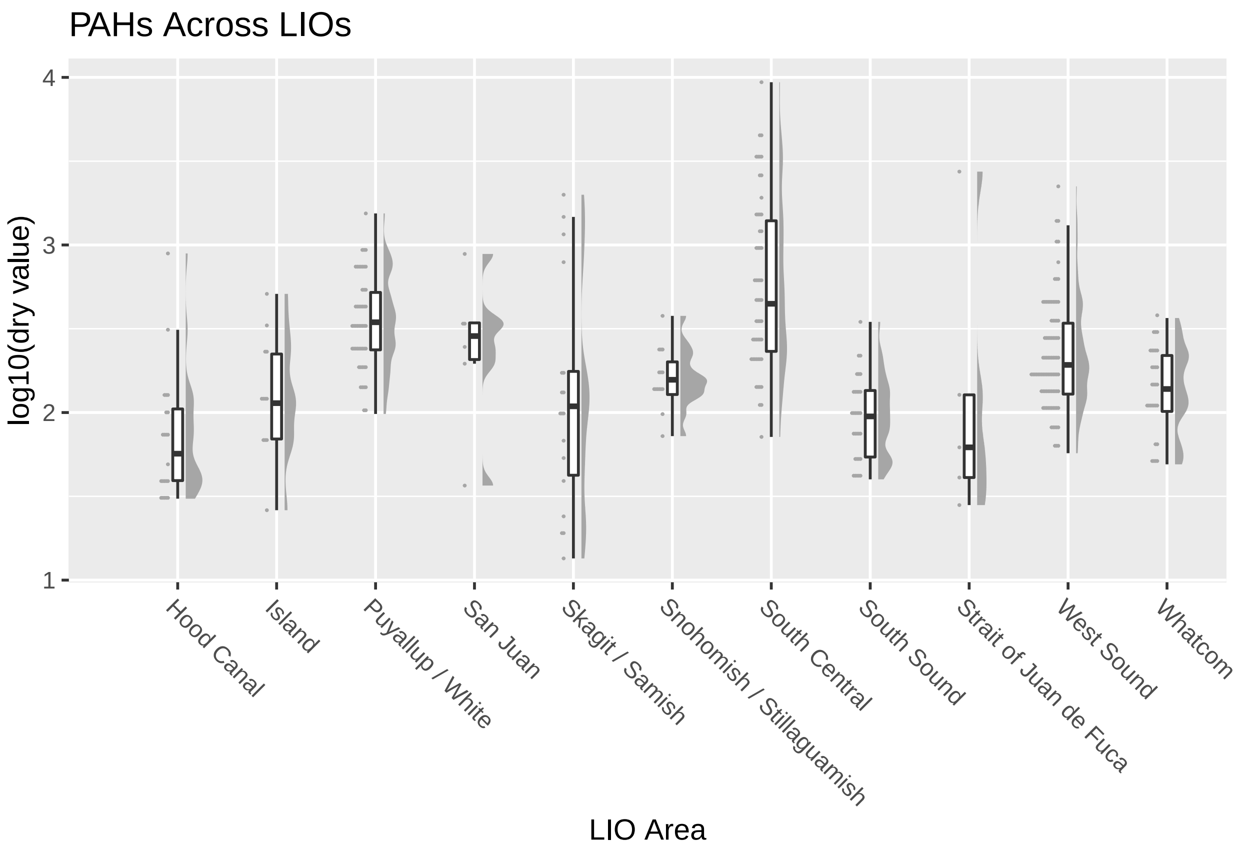


Figure 5: Distribution of the log concentration of PAHs across LIOs.

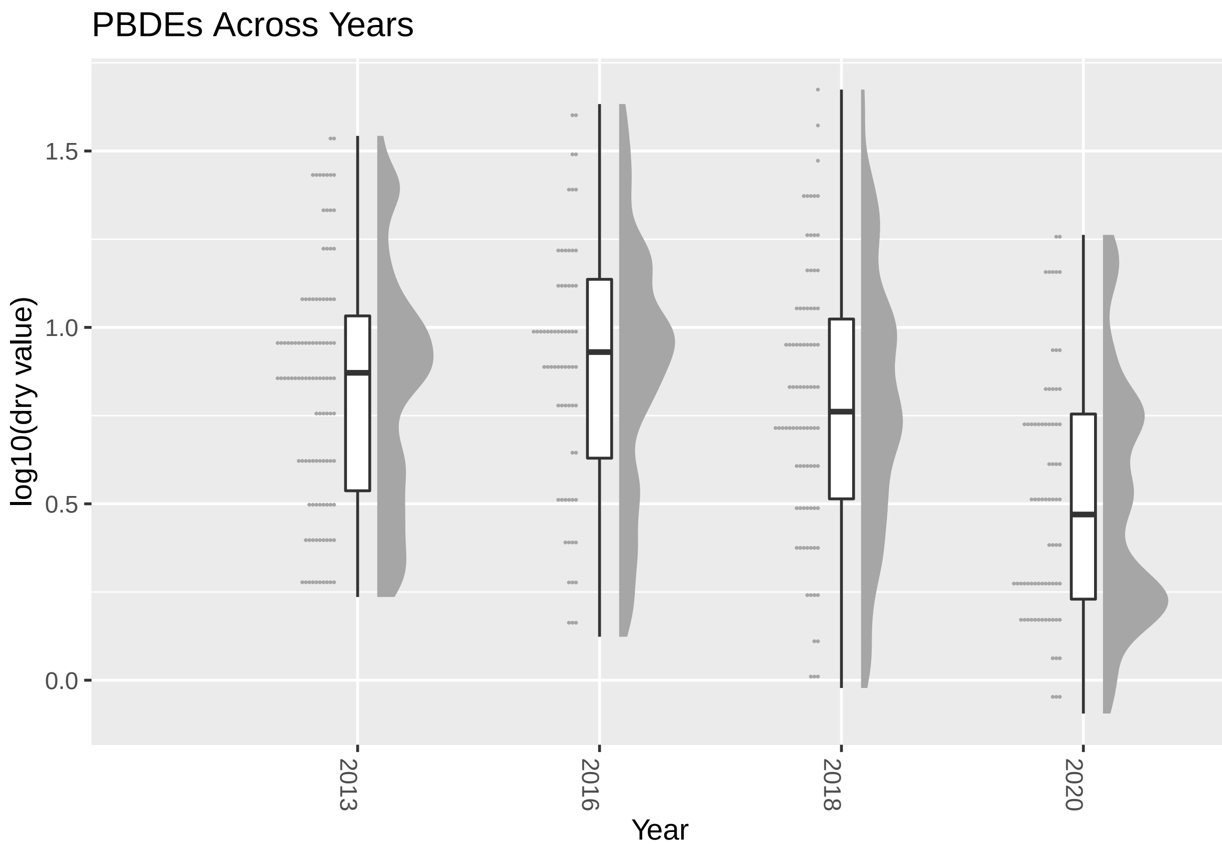


Figure 6: Distribution of the log concentration of PBDEs across years.

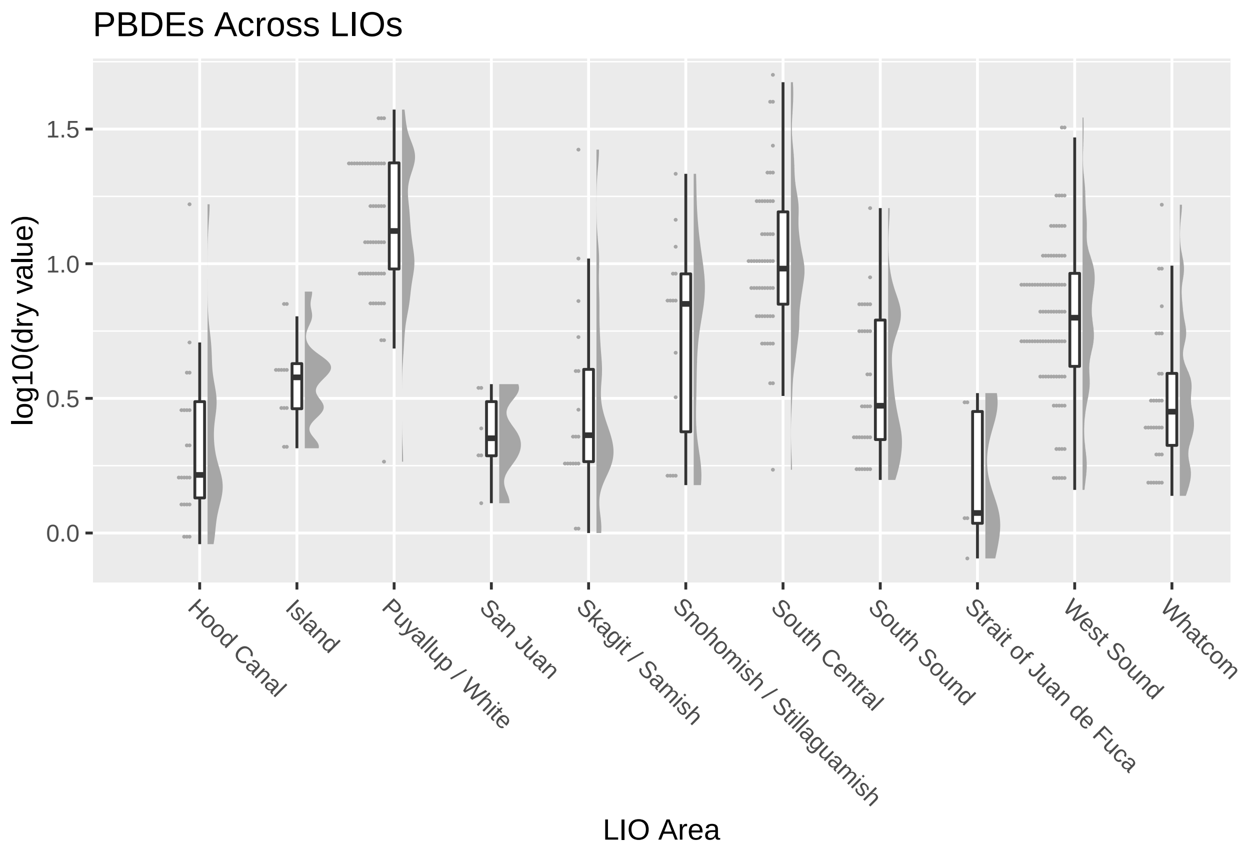


Figure 7: Distribution of the log concentration of PBDEs across LIOs.

Figure 8: Distribution of the log concentration of PCBs across years.

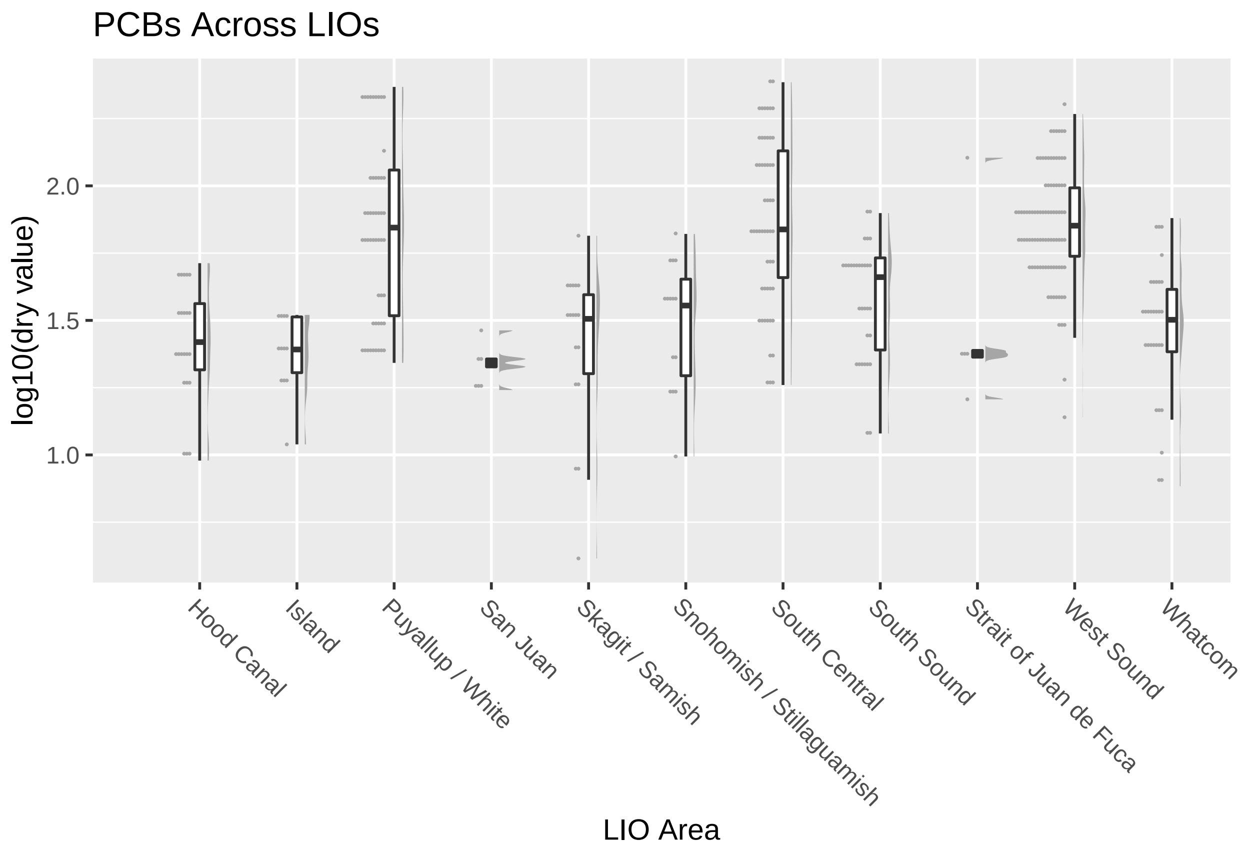


Figure 9: Distribution of the log concentration of PCBs across LIOs.

2.3 Statistical modeling

We modeled the dry weight of the analytes found in mussels using a linear mixed model (LMM). These models consist of fixed effects, which remain constant, and random effects that follow a normal distribution and can correspondingly vary by individual, location, etc. We considered the following explanatory variables in our analysis:

- time in days that the mussels spent in the water;
- latitude and longitude;
- year (as a fixed factor);
- county;
- LIO;
- WRIA;
- mean percent AU.

Other extraneous variables (IE, funding source) were not used as there is no possible relationship between these and analyte content, though this information could be helpful when we begin to focus on future sampling plans. Exploratory analysis of the data allowed us to remove extraneous factors, while including those we wished to investigate. Models were created for each of the analytes independently, as their nature and distribution should not be assumed to be the same.

Initial model selection occurred by comparing the Akaike Information Criterion (AIC) of all feasible possible models. Models that were unsuitable due to singular fits (overfitting) or violations or model assumptions such as non-normality were removed. The final model was chosen by taking into account AIC, noninfringement of model assumptions, and variables critical to project goals.

3 Results

3.1 Model selection

Based upon ΔAIC values (Tables 1-3), the model with the most support from the data for all analytes included fixed effects for

- *year*;
- a $year \times LIO$ interaction;
- mean percent impervious surface in the adjacent shoreline watershed assessment unit (AU); and
- number of *days* the mussels were immersed.

The model also included the random effect of longitude, but not latitude. There was very little evidence to support the inclusion of LIO as a random effect either.

Specifically, the form of the best supported model is

$$D_{a,i} = \alpha_{a,year} + \gamma_{a,year \times LIO} + \beta_a \text{surface}_{a,i} + \tau_a \text{time}_{a,i} + \mu_{a,[LIO:lon]} + \epsilon_{a,i} \tag{1}$$

where $D_{a,i}$ is the natural logarithm of the dry weight of analyte a from a sample site i, $\alpha_{a,year}$ is the categorical effect of year on analyte a, $\gamma_{a,year \times LIO}$ is the interaction effect of year and LIO on analyte a, β_a is the effect of mean percent impervious surface of the nearest watershed region on analyte a from location i, τ_a is the effect of the time the mussels remained in the water (which varied by sampling year) at location i for analyte a, $\mu_{a,[LIO:lon]}$ is the random effect of longitude nested within LIO on analyte a, such that $\mu_{a,[LIO:lon]} \sim \text{N}(0, \sigma_\mu^2)$, and $\epsilon_{a,i}$ is the residual error, which is normally distributed with $\epsilon_{a,i} \sim \text{N}(0, \sigma_\epsilon^2)$.

Table 1: Model selection results for PAHs.

	Random effect	ΔAIC
5	longitude	0.0
2	longitude in LIO	2.0
4	latitude	7.9
1	latitude in LIO	9.9
3	LIO	32.5

Table 2: Model selection results for PBDEs.

	Random effect	ΔAIC
5	longitude	0.0
4	latitude	0.9
2	longitude in LIO	2.0
1	latitude in LIO	2.9
3	LIO	3.1

Table 3: Model selection results for PCBs.

	Random effect	ΔAIC
5	longitude	0.0
2	longitude in LIO	2.0
4	latitude	5.5
1	latitude in LIO	7.5
3	LIO	14.1

3.2 Variable importance

Based upon model sum-of-squares, the mean percent of impervious surface explains a large proportion of the variance in PAHs, PBDEs, and PCBs (Tables 4-6). The interactive effects of year and LIO also appear important, suggesting shifts in the distributions of analyte concentrations over time.

Table 4: ANOVA table for the model of PAHs.

	df	Sum Sq	Mean Sq	F-value
year	3	1.3	0.4	7.30
time	1	0.8	0.8	13.77
mean_is_au	1	11.7	11.7	196.11
year:lio_areas	28	6.3	0.2	3.78

Table 5: ANOVA table for the model of PBDEs.

	df	Sum Sq	Mean Sq	F-value
year	3	23.8	7.9	48.16
time	1	0.1	0.1	0.47
mean_is_au	1	39.6	39.6	240.12
year:lio_areas	28	34.6	1.2	7.49

Table 6: ANOVA table for the model of PCBs.

	df	Sum Sq	Mean Sq	F-value
year	3	3.9	1.3	19.18
time	1	0.1	0.1	1.57
mean_is_au	1	6.6	6.6	98.24
year:lio_areas	28	10.0	0.4	5.29

3.3 Parameter estimates

Parameter estimates and their associate 95% confidence intervals from the best supported models are provided in the following tables. Additional information about the model parameters is provided in the Appendix.

Table 7: Parameter estimates and 95% confidence intervals for the model of PAHs.

Characteristic	**Beta**	**95% CI**
year		
2013	—	—
2016	1.1	-0.59, 2.9

Characteristic	**Beta**	**95% CI**
2018	0.65	-0.63, 1.9
2020	0.65	-0.66, 2.0
time	-0.03	-0.06, 0.00
mean_is_au	0.02	0.01, 0.02
year * lio_areas		
2013 * Puyallup / White	0.64	-0.27, 1.5
2016 * Puyallup / White	0.82	-0.08, 1.7
2018 * Puyallup / White	0.94	0.29, 1.6
2020 * Puyallup / White	0.39	-0.32, 1.1
2013 * Skagit / Samish	-0.62	-1.6, 0.36
2016 * Skagit / Samish	1.1	0.05, 2.2
2018 * Skagit / Samish	0.71	-0.17, 1.6
2020 * Skagit / Samish	0.32	-0.57, 1.2
2013 * Snohomish / Stillaguamish	0.21	-0.83, 1.3
2016 * Snohomish / Stillaguamish	0.62	-0.37, 1.6
2018 * Snohomish / Stillaguamish	0.34	-0.47, 1.2
2020 * Snohomish / Stillaguamish	0.26	-0.52, 1.0
2013 * South Central	1.0	0.12, 1.9
2016 * South Central	1.8	1.0, 2.6
2018 * South Central	1.7	1.1, 2.3
2020 * South Central	1.2	0.53, 1.8
2013 * South Sound	-0.43	-1.4, 0.50
2016 * South Sound	0.57	-0.32, 1.5
2018 * South Sound	0.23	-0.48, 0.94
2020 * South Sound	0.11	-0.68, 0.90
2013 * West Sound	0.62	-0.26, 1.5
2016 * West Sound	1.1	0.39, 1.9
2018 * West Sound	0.91	0.36, 1.5
2020 * West Sound	0.59	0.00, 1.2
2013 * Whatcom	0.05	-0.86, 1.0
2016 * Whatcom	0.75	-0.07, 1.6
2018 * Whatcom	0.53	-0.17, 1.2
2020 * Whatcom	0.25	-0.44, 0.94
longitude.sd__(Intercept)	0.68	
Residual.sd__Observation	0.24	

Table 8: Parameter estimates and 95% confidence intervals for the model of PBDEs.

Characteristic	**Beta**	**95% CI**
year		
2013	—	—
2016	-1.7	-2.9, -0.41
2018	-1.4	-2.3, -0.51
2020	-1.4	-2.3, -0.44
time	0.02	0.00, 0.04
mean_is_au	0.01	0.01, 0.01
year * lio_areas		
2013 * Puyallup / White	0.61	-0.03, 1.3
2016 * Puyallup / White	1.7	1.0, 2.3
2018 * Puyallup / White	1.8	1.3, 2.3
2020 * Puyallup / White	1.0	0.48, 1.5
2013 * Skagit / Samish	-0.72	-1.4, -0.02
2016 * Skagit / Samish	0.60	-0.17, 1.4
2018 * Skagit / Samish	0.71	0.09, 1.3
2020 * Skagit / Samish	-0.17	-0.81, 0.46
2013 * Snohomish / Stillaguamish	0.37	-0.37, 1.1
2016 * Snohomish / Stillaguamish	1.5	0.70, 2.4
2018 * Snohomish / Stillaguamish	1.5	0.79, 2.1
2020 * Snohomish / Stillaguamish	-0.27	-0.86, 0.31
2013 * South Central	0.37	-0.27, 1.0
2016 * South Central	1.8	1.2, 2.4
2018 * South Central	1.5	1.0, 1.9
2020 * South Central	0.73	0.25, 1.2
2013 * South Sound	-0.22	-0.89, 0.44
2016 * South Sound	0.76	0.12, 1.4
2018 * South Sound	0.51	0.01, 1.0
2020 * South Sound	0.10	-0.46, 0.66
2013 * West Sound	0.60	-0.03, 1.2
2016 * West Sound	1.3	0.71, 1.8
2018 * West Sound	1.1	0.70, 1.5
2020 * West Sound	0.36	-0.07, 0.80
2013 * Whatcom	-0.53	-1.2, 0.13
2016 * Whatcom	0.74	0.12, 1.4
2018 * Whatcom	0.63	0.08, 1.2
2020 * Whatcom	-0.09	-0.63, 0.45

Characteristic	Beta	95% CI
longitude.sd__(Intercept)	0.31	
Residual.sd__Observation	0.41	

Table 9: Parameter estimates and 95% confidence intervals for the model of PCBs.

Characteristic	Beta	95% CI
year		
2013	—	—
2016	0.20	-1.1, 1.5
2018	-0.17	-1.1, 0.79
2020	-0.13	-1.1, 0.85
time	0.00	-0.03, 0.02
mean_is_au	0.01	0.01, 0.01
year * lio_areas		
2013 * Puyallup / White	0.26	-0.41, 0.93
2016 * Puyallup / White	0.26	-0.41, 0.94
2018 * Puyallup / White	0.84	0.36, 1.3
2020 * Puyallup / White	0.54	0.00, 1.1
2013 * Skagit / Samish	-0.83	-1.6, -0.10
2016 * Skagit / Samish	-0.05	-0.86, 0.76
2018 * Skagit / Samish	0.30	-0.35, 0.94
2020 * Skagit / Samish	0.15	-0.51, 0.81
2013 * Snohomish / Stillaguamish	-0.19	-1.0, 0.58
2016 * Snohomish / Stillaguamish	0.40	-0.39, 1.2
2018 * Snohomish / Stillaguamish	0.58	-0.06, 1.2
2020 * Snohomish / Stillaguamish	-0.50	-1.1, 0.09
2013 * South Central	0.08	-0.58, 0.75
2016 * South Central	0.74	0.13, 1.4
2018 * South Central	1.0	0.58, 1.5
2020 * South Central	0.78	0.30, 1.3
2013 * South Sound	-0.38	-1.1, 0.31
2016 * South Sound	0.35	-0.31, 1.0
2018 * South Sound	0.71	0.19, 1.2
2020 * South Sound	0.33	-0.26, 0.91
2013 * West Sound	0.39	-0.26, 1.0
2016 * West Sound	0.70	0.13, 1.3
2018 * West Sound	1.1	0.72, 1.6

Characteristic	Beta	95% CI
2020 * West Sound	0.91	0.46, 1.4
2013 * Whatcom	-0.40	-1.1, 0.29
2016 * Whatcom	-0.08	-0.71, 0.55
2018 * Whatcom	0.35	-0.20, 0.90
2020 * Whatcom	-0.13	-0.67, 0.41
longitude.sd___(Intercept)	0.47	
Residual.sd___Observation	0.26	

3.4 Model diagnostics

In general, models met the assumptions of well-behaved, independent, and identical errors (Figures 10 - 12), but there was some indication of remaining information in the residuals for the model of PAHs (Figure 10). Although there was some indication of heavy-tailed (leptokurtic) residuals, the Q-Q plots looked quite reasonable overall.

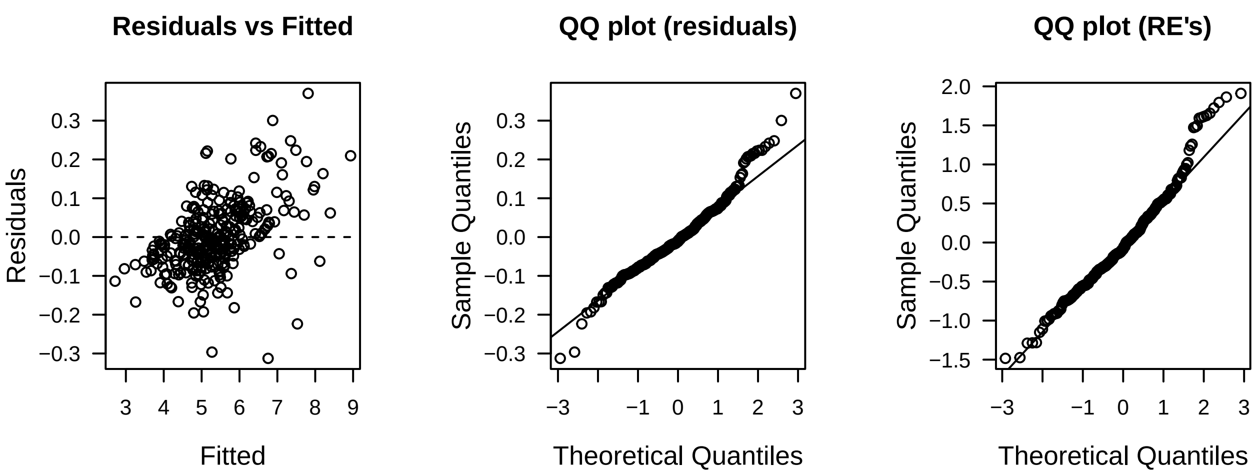


Figure 10: Model diagnostics for the top ranked model for PAHs.

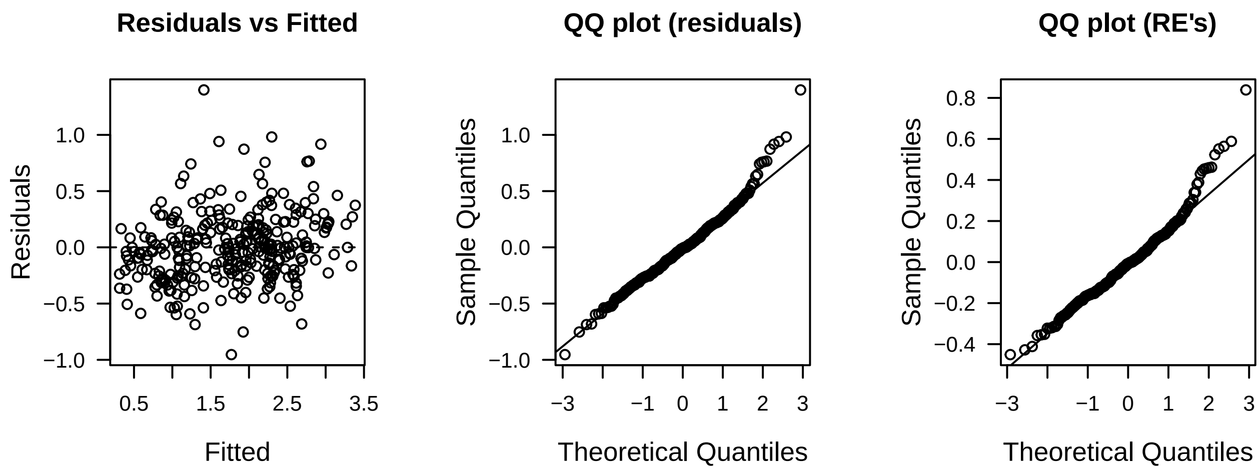


Figure 11: Model diagnostics for the top ranked model for PBDEs.

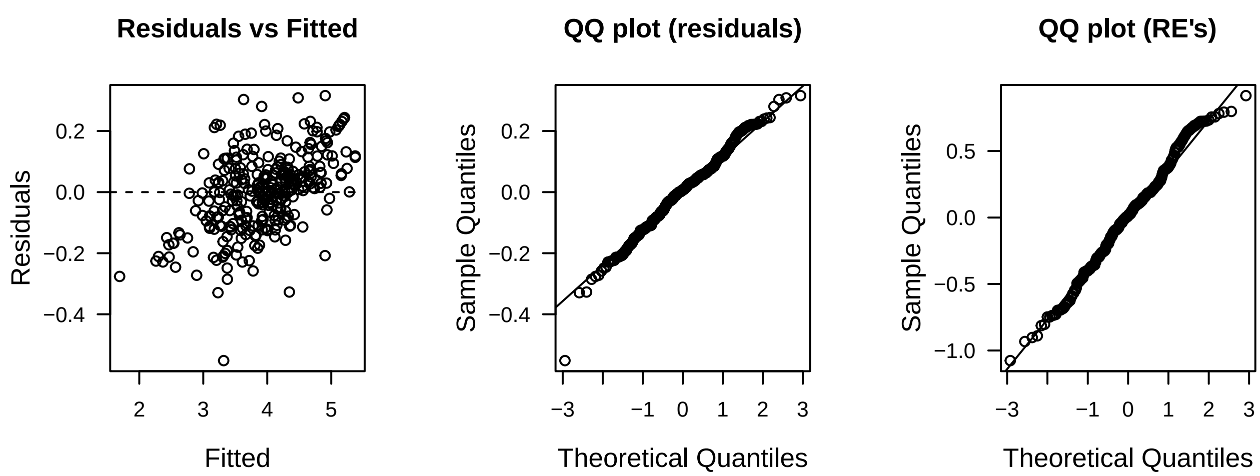


Figure 12: Model diagnostics for the top ranked model for PCBs.

3.5 Power analyses

The best supported models are near singular with respect to estimating the interaction of year and LIO, and this confounds our ability to assess the power to detect meaningful changes across years. To address this, we removed the interaction term from each model and refit them before estimating power. After doing so, the power to detect differences among years is very high (95-100%), but importantly, this should not be confounded with the power to detect long-term changes over time *per se*. That is, in this case the high power indicates that one could estimate the difference between any two years, whether or not they occurred sequentially in time.

3.5.1 PAH

Here is the power calculation for `year`.

```
## Warning in observedPowerWarning(sim): This appears to be an "observed power"
## calculation
```

```
## Power for predictor 'year', (95% confidence interval):
##       100.0% (96.38, 100.0)
##
## Test: Likelihood ratio
##
## Based on 100 simulations, (0 warnings, 0 errors)
## alpha = 0.05, nrow = 308
##
## Time elapsed: 0 h 0 m 8 s
##
## nb: result might be an observed power calculation
```

3.5.2 PBDE

Here is the power calculation for `year`.

```
## Warning in observedPowerWarning(sim): This appears to be an "observed power"
## calculation
```

```
## Power for predictor 'year', (95% confidence interval):
##       100.0% (96.38, 100.0)
##
## Test: Likelihood ratio
##
## Based on 100 simulations, (2 warnings, 0 errors)
## alpha = 0.05, nrow = 308
##
## Time elapsed: 0 h 0 m 8 s
##
## nb: result might be an observed power calculation
```

3.5.3 PCB

Here is the power calculation for `year`.

```
## Warning in observedPowerWarning(sim): This appears to be an "observed power"
## calculation
```

```
## Power for predictor 'year', (95% confidence interval):
##       95.00% (88.72, 98.36)
##
## Test: Likelihood ratio
##
## Based on 100 simulations, (0 warnings, 0 errors)
## alpha = 0.05, nrow = 308
##
## Time elapsed: 0 h 0 m 9 s
##
## nb: result might be an observed power calculation
```

4 Summary

Our modeling suggests that the mean percent of impervious surface explains a large proportion of the variance in PAHs, PBDEs, and PCBs. The interactive effects of year and LIO also appear important, suggesting shifts in the distributions of analyte concentrations over time. However, the best supported models have limited power to detect meaningful changes in analytes over time.

When we fit reduced forms of the models that omitted the important interaction of year and LIO, we found that there is very high power to detect differences among any two years in the data set, but that it does not equate to an ability to detect long-term changes across years.

5 Acknowledgments

We acknowledge the help and support from the environmental toxicology group at WDFW, including Jim West, Sandra O'Neill, Louisa Harding, and Mariko Langness.

6 Appendix

6.1 Parameter summary tables

Table 10: Parameter estimates, associated standard errors, and t-statistics for the model of PAHs.

effect	group	term	estimate	std.error	statistic
fixed	NA	(Intercept)	6.28	0.95	6.62
fixed	NA	year2016	1.14	0.89	1.29
fixed	NA	year2018	0.65	0.65	1.00
fixed	NA	year2020	0.65	0.67	0.98
fixed	NA	time	-0.03	0.02	-2.22
fixed	NA	mean_is_au	0.02	0.00	7.99
fixed	NA	year2013:lio_areasPuyallup / White	0.64	0.46	1.38
fixed	NA	year2016:lio_areasPuyallup / White	0.82	0.46	1.78
fixed	NA	year2018:lio_areasPuyallup / White	0.94	0.33	2.86
fixed	NA	year2020:lio_areasPuyallup / White	0.39	0.36	1.08
fixed	NA	year2013:lio_areasSkagit / Samish	-0.62	0.50	-1.23
fixed	NA	year2016:lio_areasSkagit / Samish	1.14	0.55	2.05
fixed	NA	year2018:lio_areasSkagit / Samish	0.71	0.44	1.59
fixed	NA	year2020:lio_areasSkagit / Samish	0.32	0.45	0.70
fixed	NA	year2013:lio_areasSnohomish / Stillaguamish	0.21	0.53	0.40
fixed	NA	year2016:lio_areasSnohomish / Stillaguamish	0.62	0.50	1.23
fixed	NA	year2018:lio_areasSnohomish / Stillaguamish	0.34	0.41	0.82
fixed	NA	year2020:lio_areasSnohomish / Stillaguamish	0.26	0.40	0.65
fixed	NA	year2013:lio_areasSouth Central	1.01	0.45	2.24
fixed	NA	year2016:lio_areasSouth Central	1.79	0.41	4.41
fixed	NA	year2018:lio_areasSouth Central	1.71	0.31	5.51
fixed	NA	year2020:lio_areasSouth Central	1.16	0.32	3.58
fixed	NA	year2013:lio_areasSouth Sound	-0.43	0.48	-0.91
fixed	NA	year2016:lio_areasSouth Sound	0.57	0.45	1.25
fixed	NA	year2018:lio_areasSouth Sound	0.23	0.36	0.64
fixed	NA	year2020:lio_areasSouth Sound	0.11	0.40	0.27
fixed	NA	year2013:lio_areasWest Sound	0.62	0.45	1.39
fixed	NA	year2016:lio_areasWest Sound	1.14	0.38	2.98

effect	group	term	estimate	std.error	statistic
fixed	NA	year2018:lio_areasWest Sound	0.91	0.28	3.24
fixed	NA	year2020:lio_areasWest Sound	0.59	0.30	1.95
fixed	NA	year2013:lio_areasWhatcom	0.05	0.46	0.10
fixed	NA	year2016:lio_areasWhatcom	0.75	0.42	1.79
fixed	NA	year2018:lio_areasWhatcom	0.53	0.36	1.48
fixed	NA	year2020:lio_areasWhatcom	0.25	0.35	0.71
ran_pars	longitude	sd___(Intercept)	0.68	NA	NA
ran_pars	Residual	sd___Observation	0.24	NA	NA

Table 11: Parameter estimates, associated standard errors, and t-statistics for the model of PBDEs.

effect	group	term	estimate	std.error	statistic
fixed	NA	(Intercept)	0.35	0.68	0.51
fixed	NA	year2016	-1.66	0.64	-2.61
fixed	NA	year2018	-1.43	0.47	-3.05
fixed	NA	year2020	-1.38	0.48	-2.88
fixed	NA	time	0.02	0.01	1.68
fixed	NA	mean_is_au	0.01	0.00	7.14
fixed	NA	year2013:lio_areasPuyallup / White	0.61	0.33	1.86
fixed	NA	year2016:lio_areasPuyallup / White	1.65	0.33	5.04
fixed	NA	year2018:lio_areasPuyallup / White	1.78	0.24	7.43
fixed	NA	year2020:lio_areasPuyallup / White	1.01	0.27	3.74
fixed	NA	year2013:lio_areasSkagit / Samish	-0.72	0.36	-2.01
fixed	NA	year2016:lio_areasSkagit / Samish	0.60	0.40	1.53
fixed	NA	year2018:lio_areasSkagit / Samish	0.71	0.32	2.24
fixed	NA	year2020:lio_areasSkagit / Samish	-0.17	0.32	-0.54
fixed	NA	year2013:lio_areasSnohomish / Stillaguamish	0.37	0.38	0.98
fixed	NA	year2016:lio_areasSnohomish / Stillaguamish	1.55	0.43	3.58
fixed	NA	year2018:lio_areasSnohomish / Stillaguamish	1.46	0.34	4.26
fixed	NA	year2020:lio_areasSnohomish / Stillaguamish	-0.27	0.30	-0.91
fixed	NA	year2013:lio_areasSouth Central	0.37	0.33	1.13
fixed	NA	year2016:lio_areasSouth Central	1.81	0.31	5.79
fixed	NA	year2018:lio_areasSouth Central	1.46	0.23	6.29

effect	group	term	estimate	std.error	statistic
fixed	NA	year2020:lio_areasSouth Central	0.73	0.25	2.96
fixed	NA	year2013:lio_areasSouth Sound	-0.22	0.34	-0.66
fixed	NA	year2016:lio_areasSouth Sound	0.76	0.32	2.34
fixed	NA	year2018:lio_areasSouth Sound	0.51	0.26	1.99
fixed	NA	year2020:lio_areasSouth Sound	0.10	0.29	0.34
fixed	NA	year2013:lio_areasWest Sound	0.60	0.32	1.87
fixed	NA	year2016:lio_areasWest Sound	1.26	0.28	4.52
fixed	NA	year2018:lio_areasWest Sound	1.11	0.21	5.30
fixed	NA	year2020:lio_areasWest Sound	0.36	0.22	1.63
fixed	NA	year2013:lio_areasWhatcom	-0.53	0.34	-1.57
fixed	NA	year2016:lio_areasWhatcom	0.74	0.32	2.32
fixed	NA	year2018:lio_areasWhatcom	0.63	0.28	2.26
fixed	NA	year2020:lio_areasWhatcom	-0.09	0.27	-0.33
ran_pars	longitude	sd___(Intercept)	0.31	NA	NA
ran_pars	Residual	sd___Observation	0.41	NA	NA

Table 12: Parameter estimates, associated standard errors, and t-statistics for the model of PCBs.

effect	group	term	estimate	std.error	statistic
fixed	NA	(Intercept)	3.51	0.71	4.94
fixed	NA	year2016	0.20	0.66	0.31
fixed	NA	year2018	-0.17	0.49	-0.34
fixed	NA	year2020	-0.13	0.50	-0.25
fixed	NA	time	0.00	0.01	-0.28
fixed	NA	mean_is_au	0.01	0.00	5.97
fixed	NA	year2013:lio_areasPuyallup / White	0.26	0.34	0.76
fixed	NA	year2016:lio_areasPuyallup / White	0.26	0.34	0.77
fixed	NA	year2018:lio_areasPuyallup / White	0.84	0.25	3.40
fixed	NA	year2020:lio_areasPuyallup / White	0.54	0.28	1.95
fixed	NA	year2013:lio_areasSkagit / Samish	-0.83	0.37	-2.24
fixed	NA	year2016:lio_areasSkagit / Samish	-0.05	0.41	-0.12
fixed	NA	year2018:lio_areasSkagit / Samish	0.30	0.33	0.90
fixed	NA	year2020:lio_areasSkagit / Samish	0.15	0.34	0.44
fixed	NA	year2013:lio_areasSnohomish / Stillaguamish	-0.19	0.39	-0.49
fixed	NA	year2016:lio_areasSnohomish / Stillaguamish	0.40	0.40	0.99

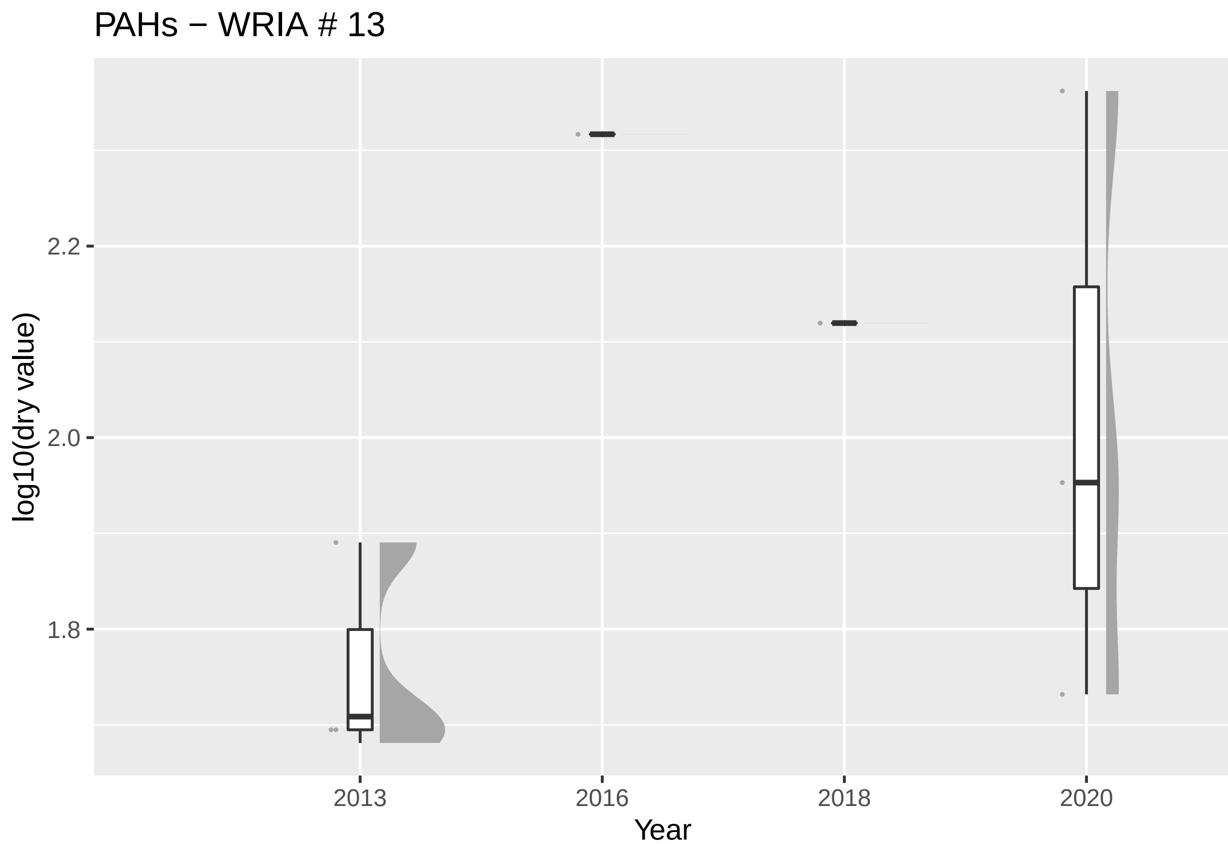
effect	group	term	estimate	std.error	statistic
fixed	NA	year2018:lio_areasSnohomish / Stillaguamish	0.58	0.33	1.77
fixed	NA	year2020:lio_areasSnohomish / Stillaguamish	-0.50	0.30	-1.65
fixed	NA	year2013:lio_areasSouth Central	0.08	0.34	0.25
fixed	NA	year2016:lio_areasSouth Central	0.74	0.31	2.38
fixed	NA	year2018:lio_areasSouth Central	1.05	0.24	4.42
fixed	NA	year2020:lio_areasSouth Central	0.78	0.25	3.16
fixed	NA	year2013:lio_areasSouth Sound	-0.38	0.35	-1.08
fixed	NA	year2016:lio_areasSouth Sound	0.35	0.34	1.03
fixed	NA	year2018:lio_areasSouth Sound	0.71	0.27	2.66
fixed	NA	year2020:lio_areasSouth Sound	0.33	0.30	1.10
fixed	NA	year2013:lio_areasWest Sound	0.39	0.33	1.18
fixed	NA	year2016:lio_areasWest Sound	0.70	0.29	2.41
fixed	NA	year2018:lio_areasWest Sound	1.14	0.21	5.34
fixed	NA	year2020:lio_areasWest Sound	0.91	0.23	3.96
fixed	NA	year2013:lio_areasWhatcom	-0.40	0.35	-1.14
fixed	NA	year2016:lio_areasWhatcom	-0.08	0.32	-0.25
fixed	NA	year2018:lio_areasWhatcom	0.35	0.28	1.25
fixed	NA	year2020:lio_areasWhatcom	-0.13	0.27	-0.46
ran_pars	longitude	sd___(Intercept)	0.47	NA	NA
ran_pars	Residual	sd___Observation	0.26	NA	NA

6.2 Raincloud plots

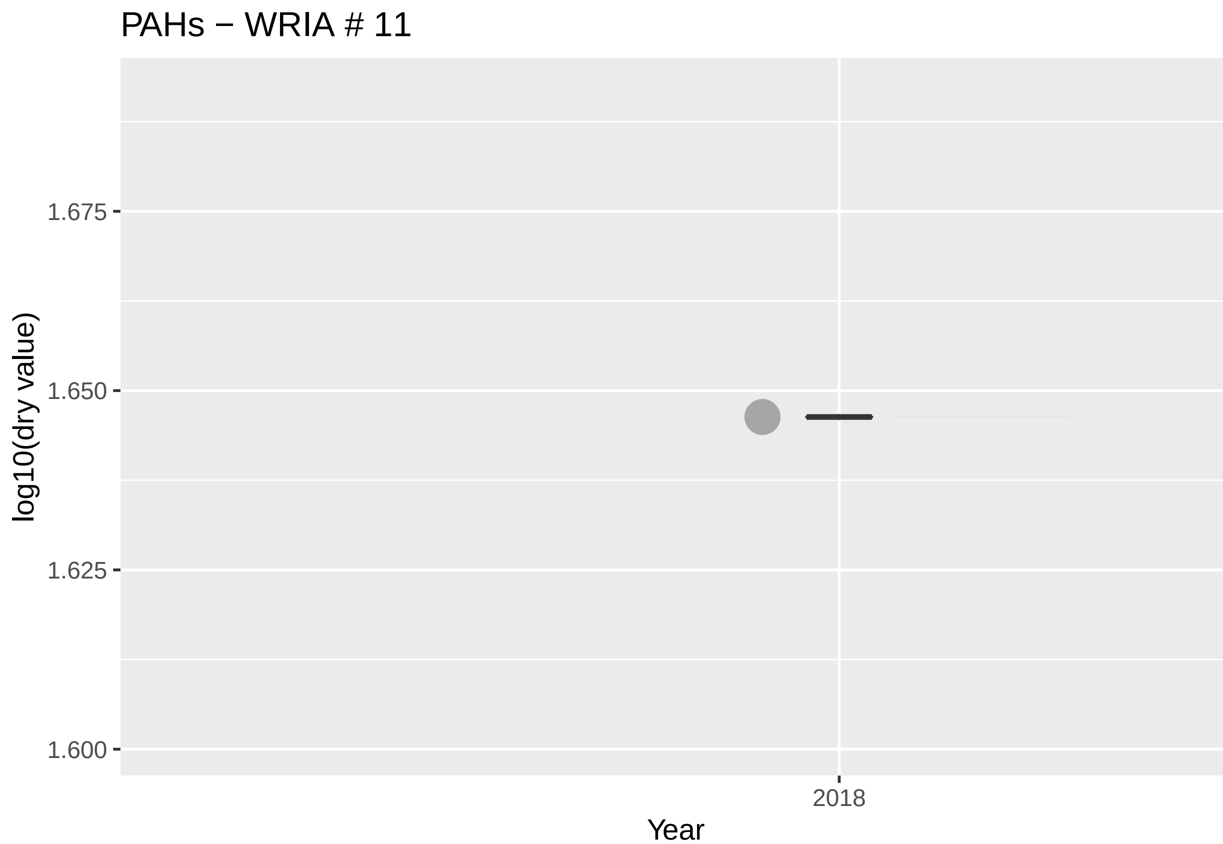
Here are additional plots of the distribution of analytes across WRIA and time.

6.2.1 PAH

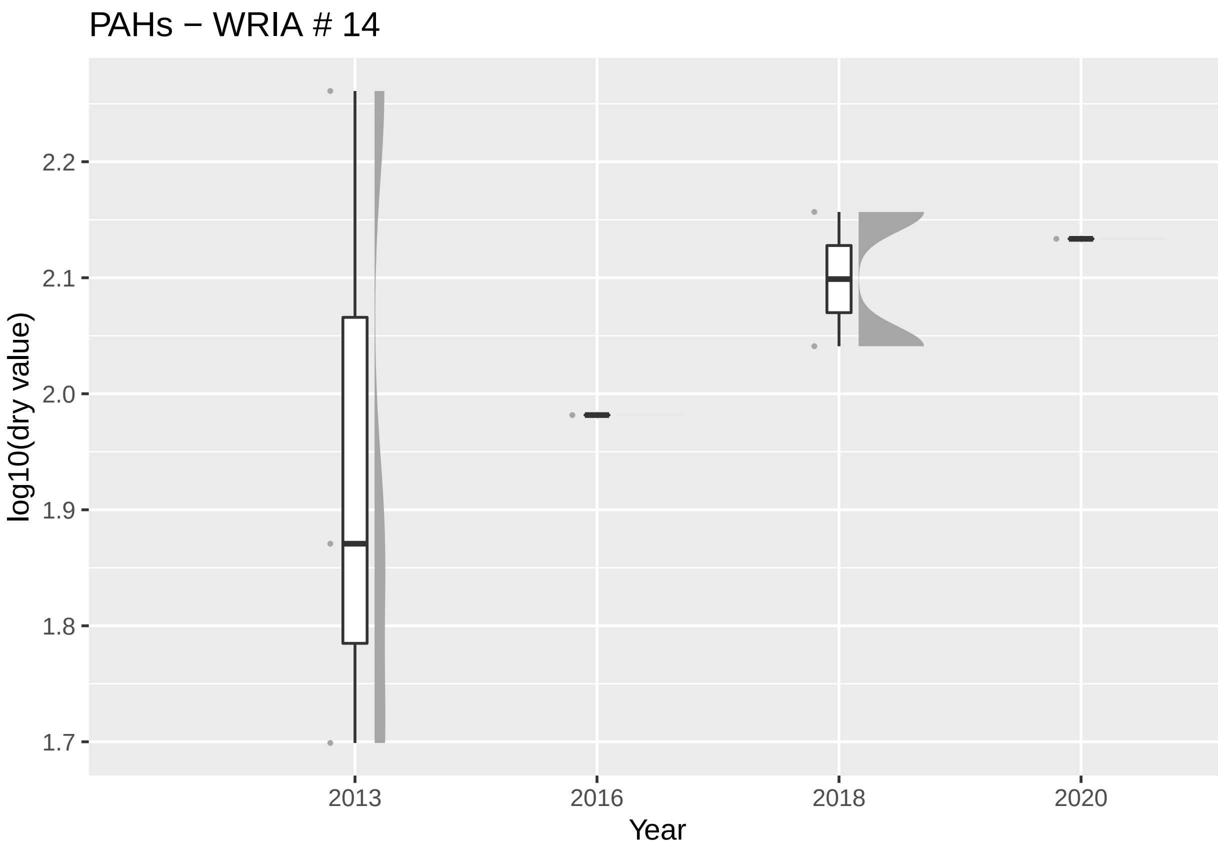
```
## $‘13‘
```



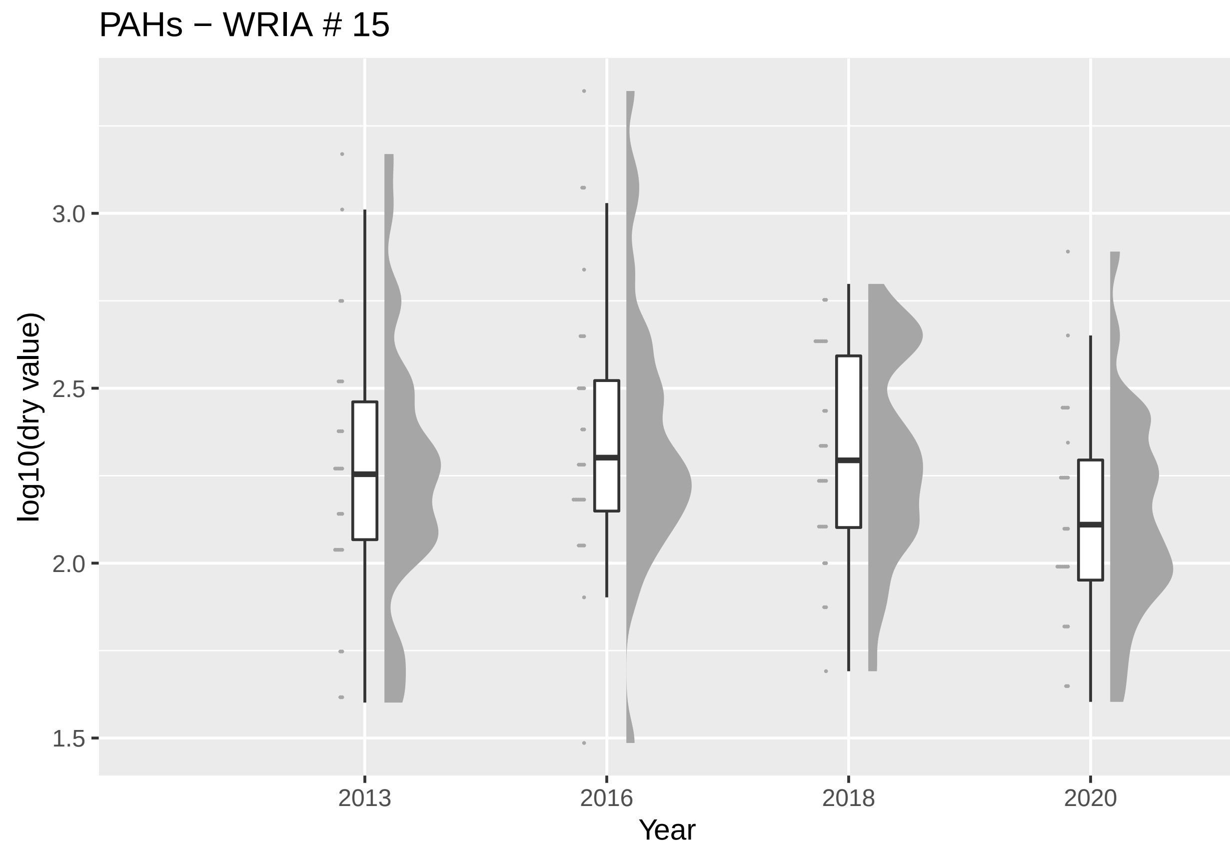

```
##
## $`11`
```



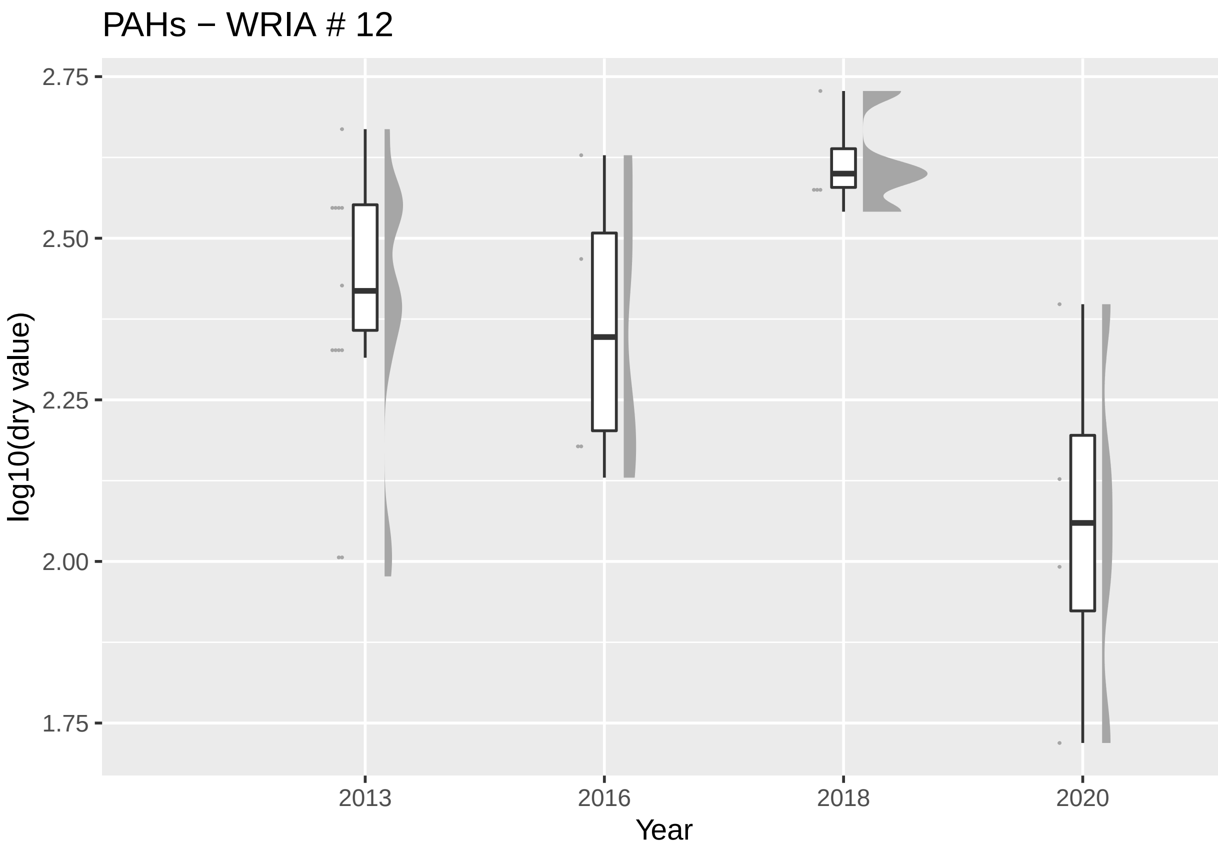

```
##
## $‘14‘
```


PAHs − WRIA # 14
log10(dry value)
2.2
2.1
2.0
1.9
1.8
1.7
2013
2016
2018
2020
Year

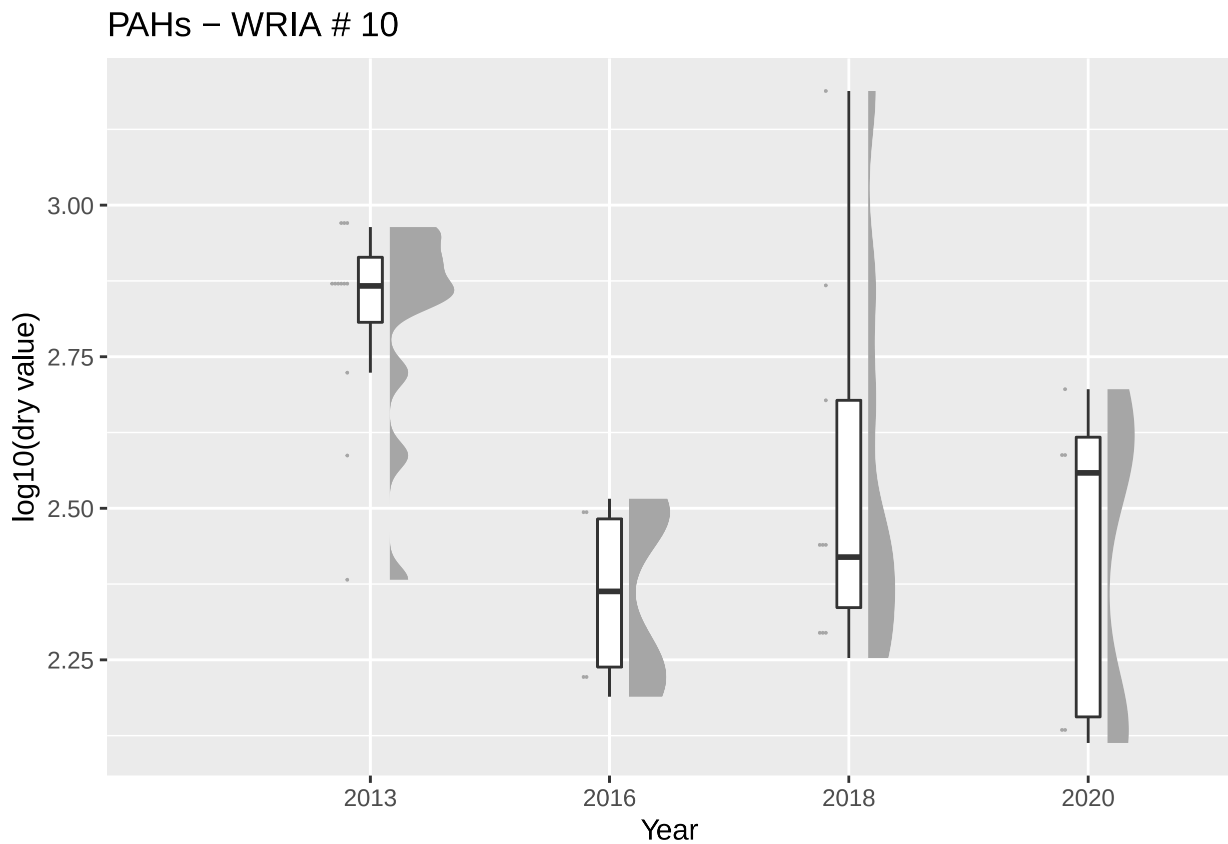
```
##
## $‘15‘
```


PAHs − WRIA # 15
log10(dry value)
3.0
2.5
2.0
1.5
2013
2016
2018
2020
Year

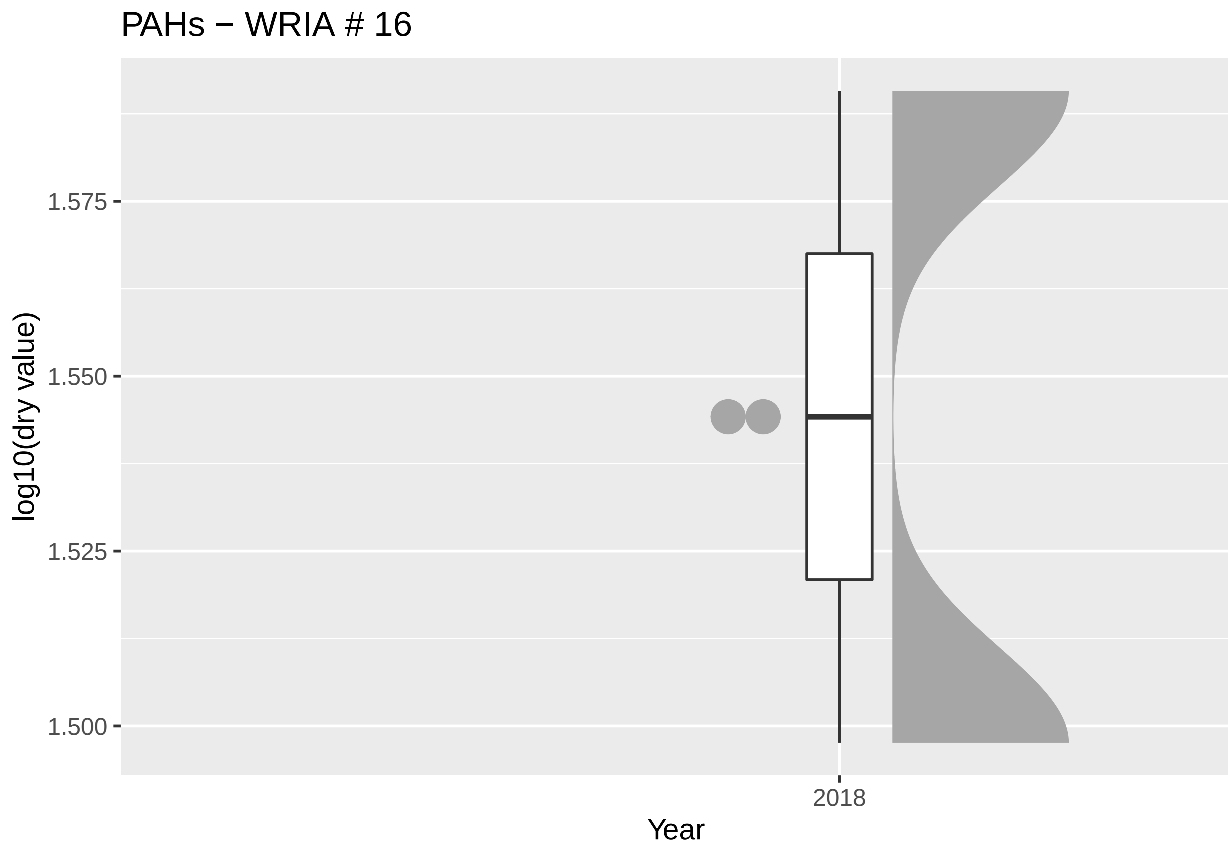
```
##
## $`12`
```


PAHs − WRIA # 12
log10(dry value)
2.75
2.50
2.25
2.00
1.75
2013
2016
2018
2020
Year

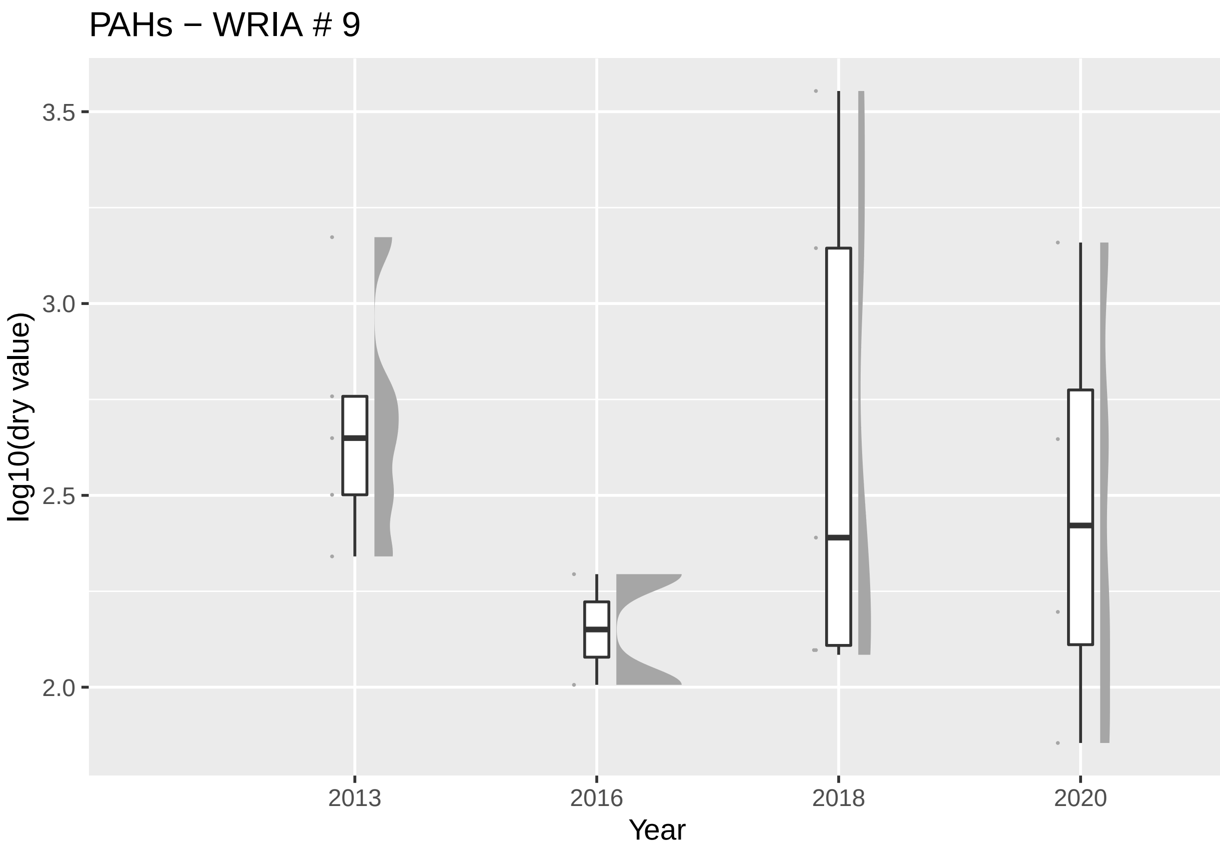
```
##
## $`10`
```


PAHs − WRIA # 10
log10(dry value)
3.00
2.75
2.50
2.25
2013
2016
2018
2020
Year

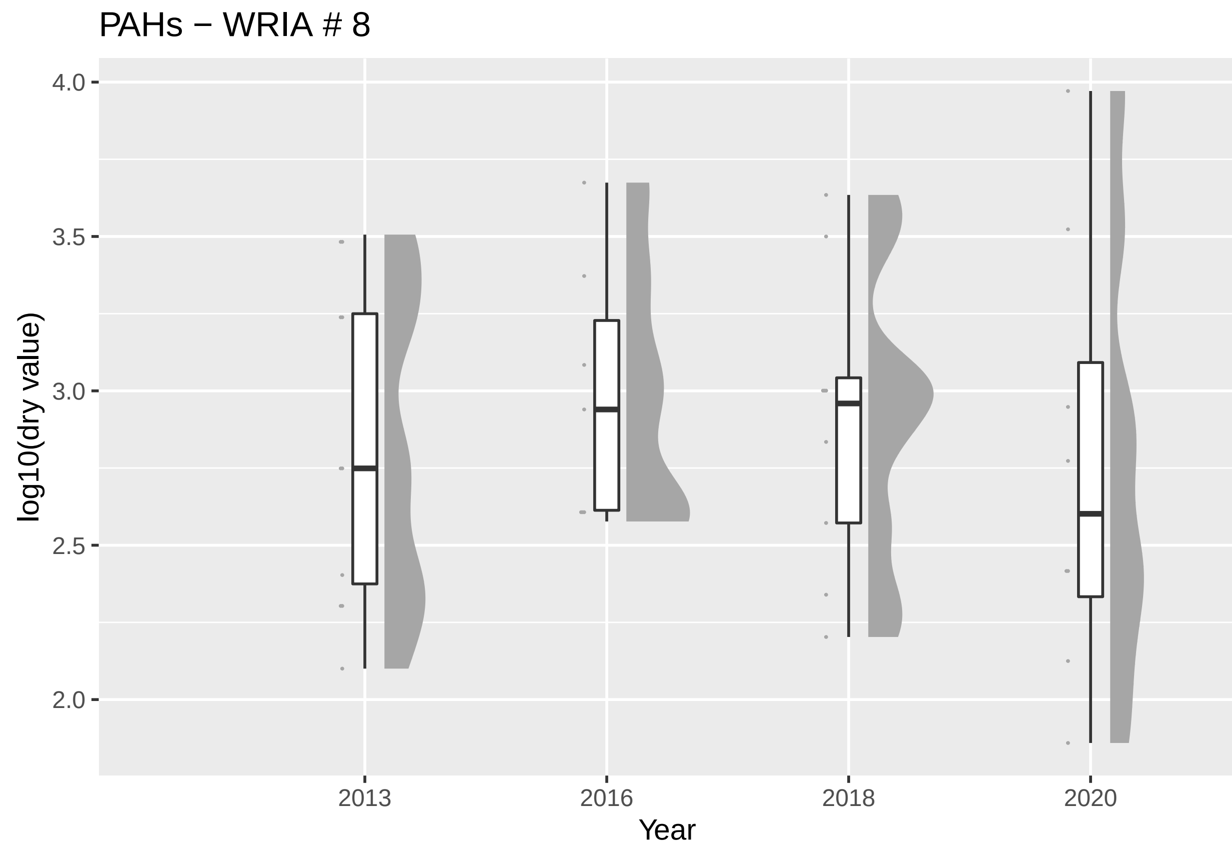
```
##
## $‘16‘
```



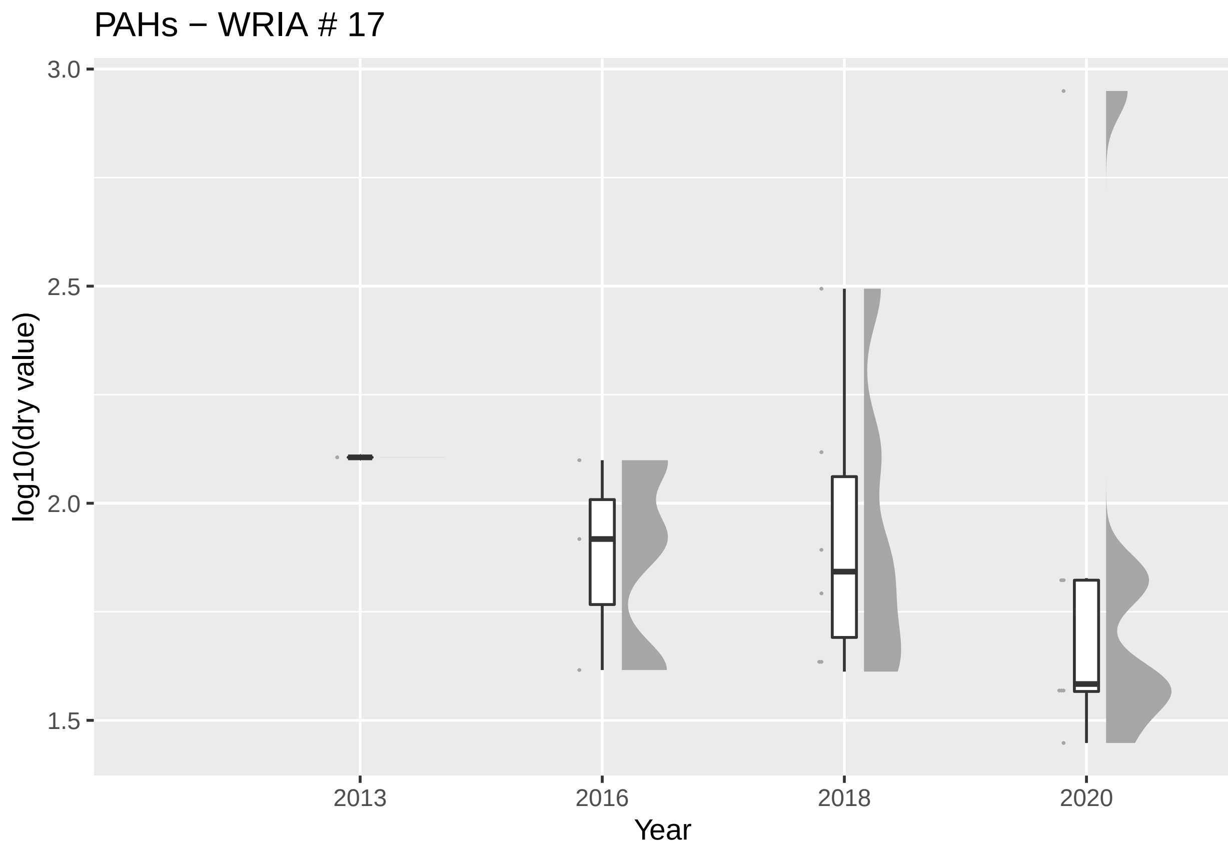

```
##
## $‘9‘
```


PAHs − WRIA # 9
log10(dry value)
3.5
3.0
2.5
2.0
2013
2016
2018
2020
Year

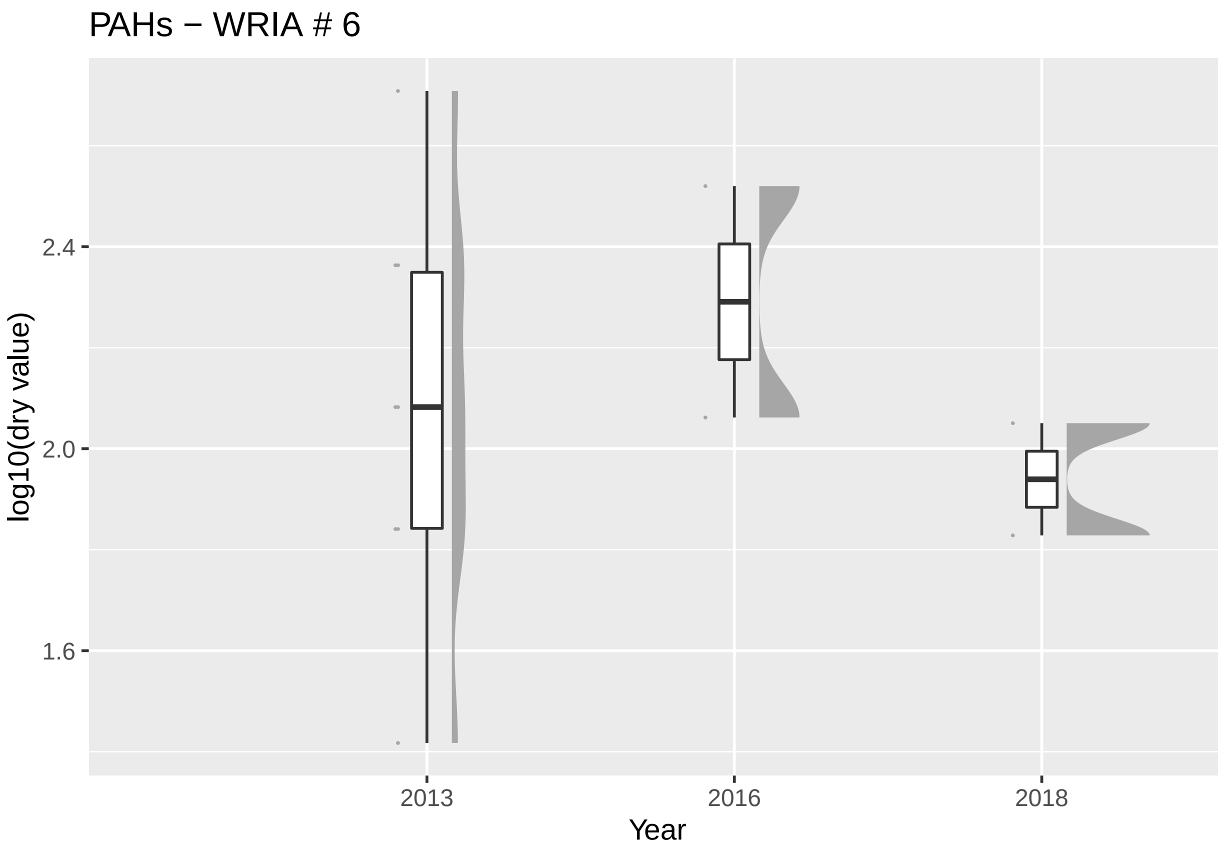
```
##
## $‘8‘
```


PAHs − WRIA # 8
4.0
3.5
3.0
2.5
2.0
log10(dry value)
2013
2016
2018
2020
Year

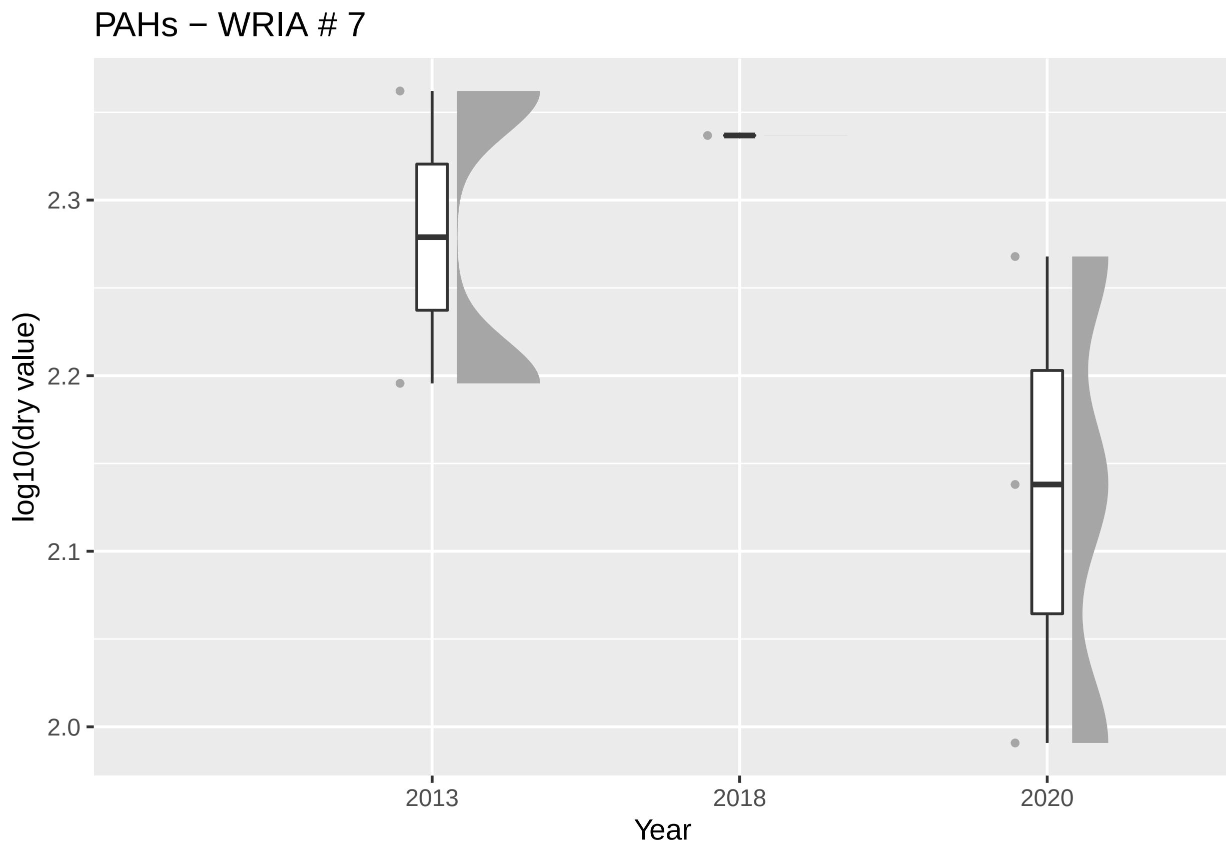
```
##
## $‘17‘
```


PAHs − WRIA # 17
log10(dry value)
3.0
2.5
2.0
1.5
2013
2016
2018
2020
Year

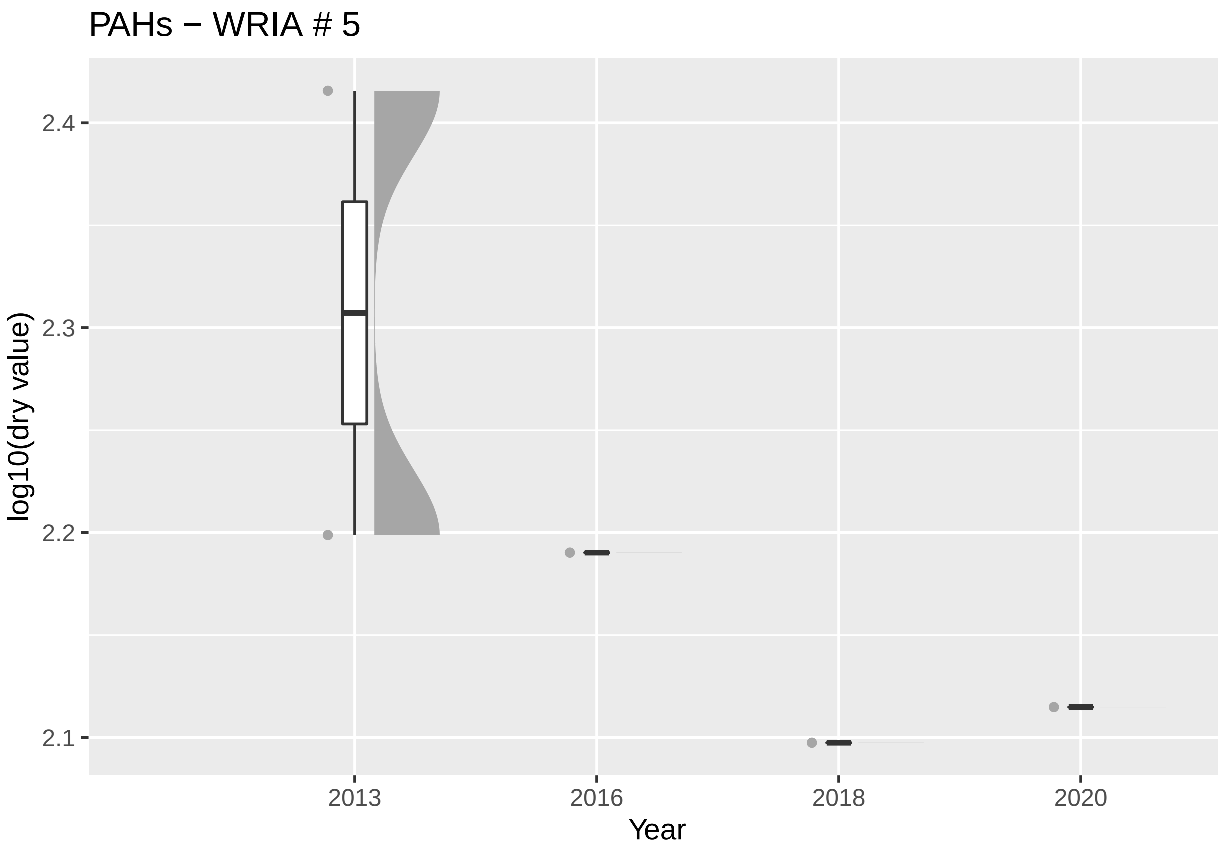
```
##
## $‘6‘
```



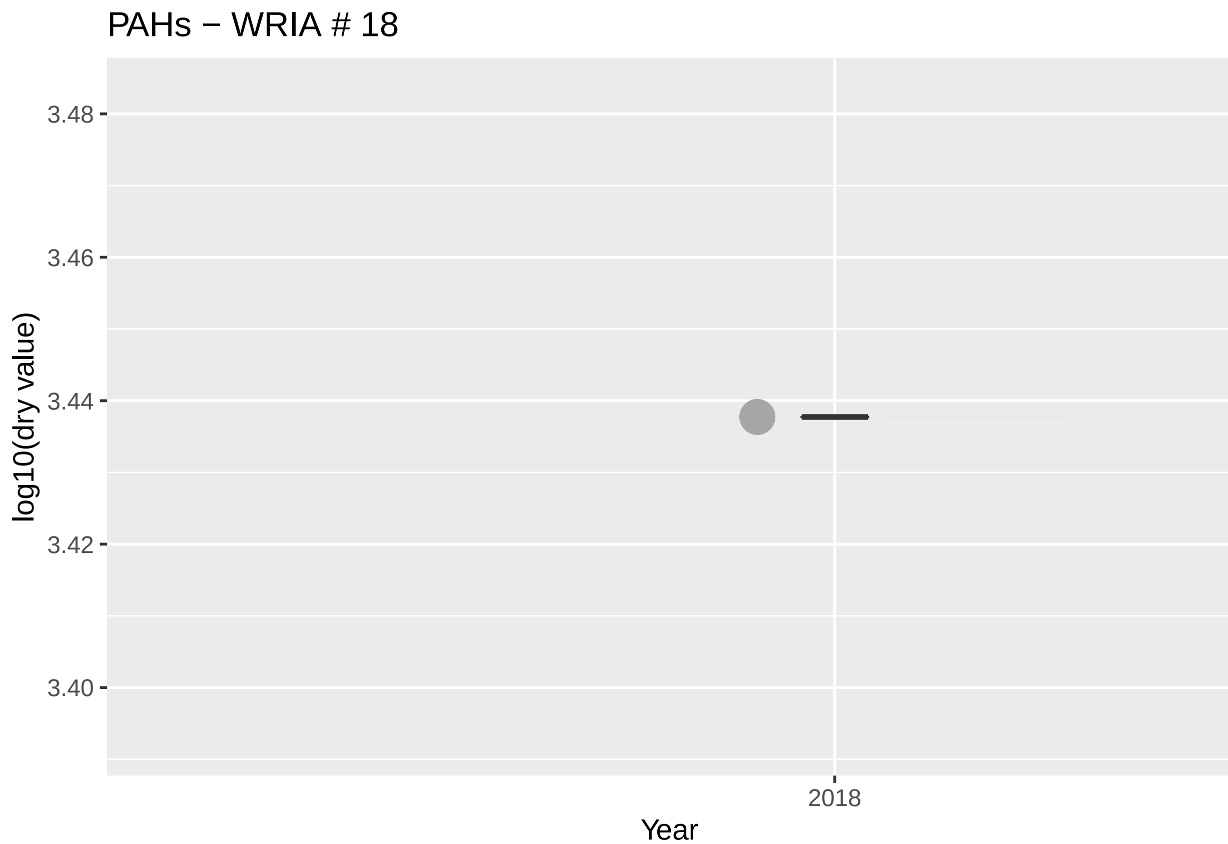

```
##
## $‘7‘
```



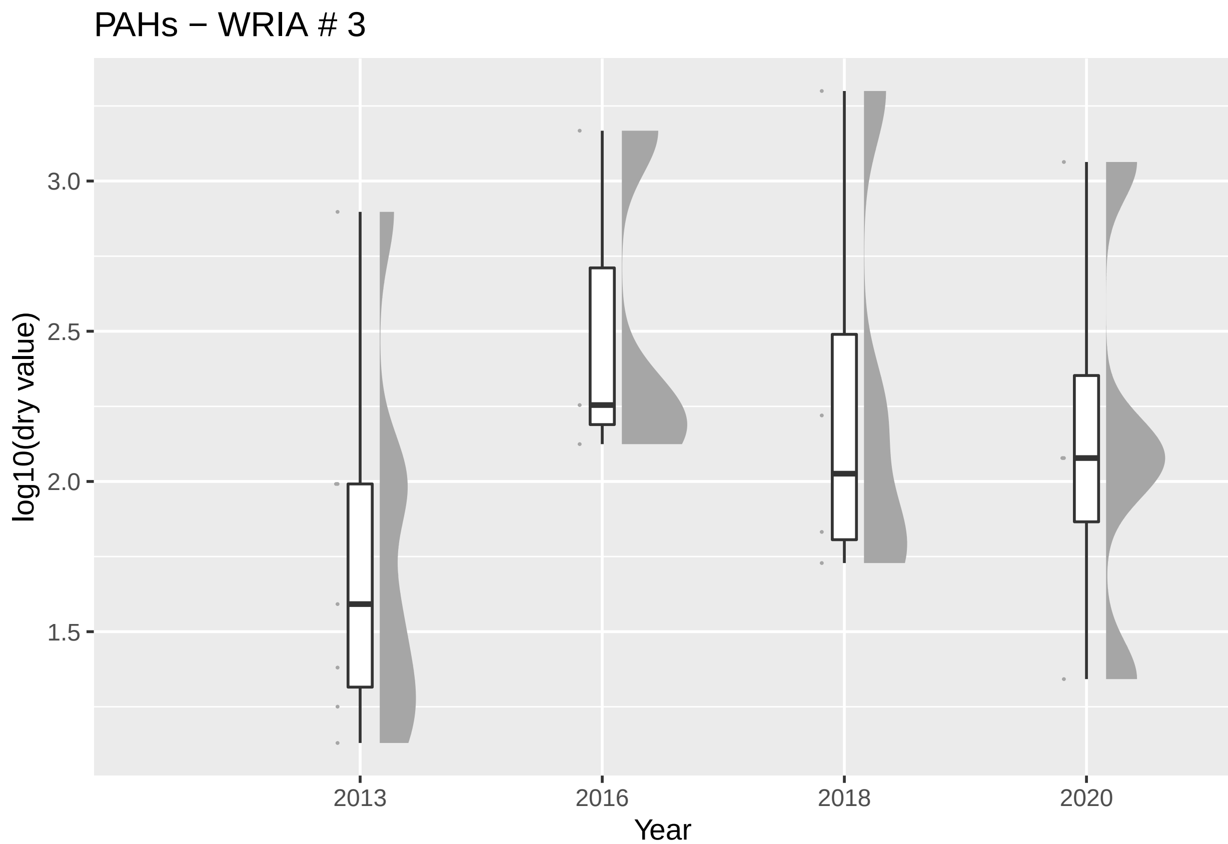

```
##
## $‘5‘
```


PAHs − WRIA # 5
2.4
2.3
2.2
2.1
log10(dry value)
2013
2016
2018
2020
Year

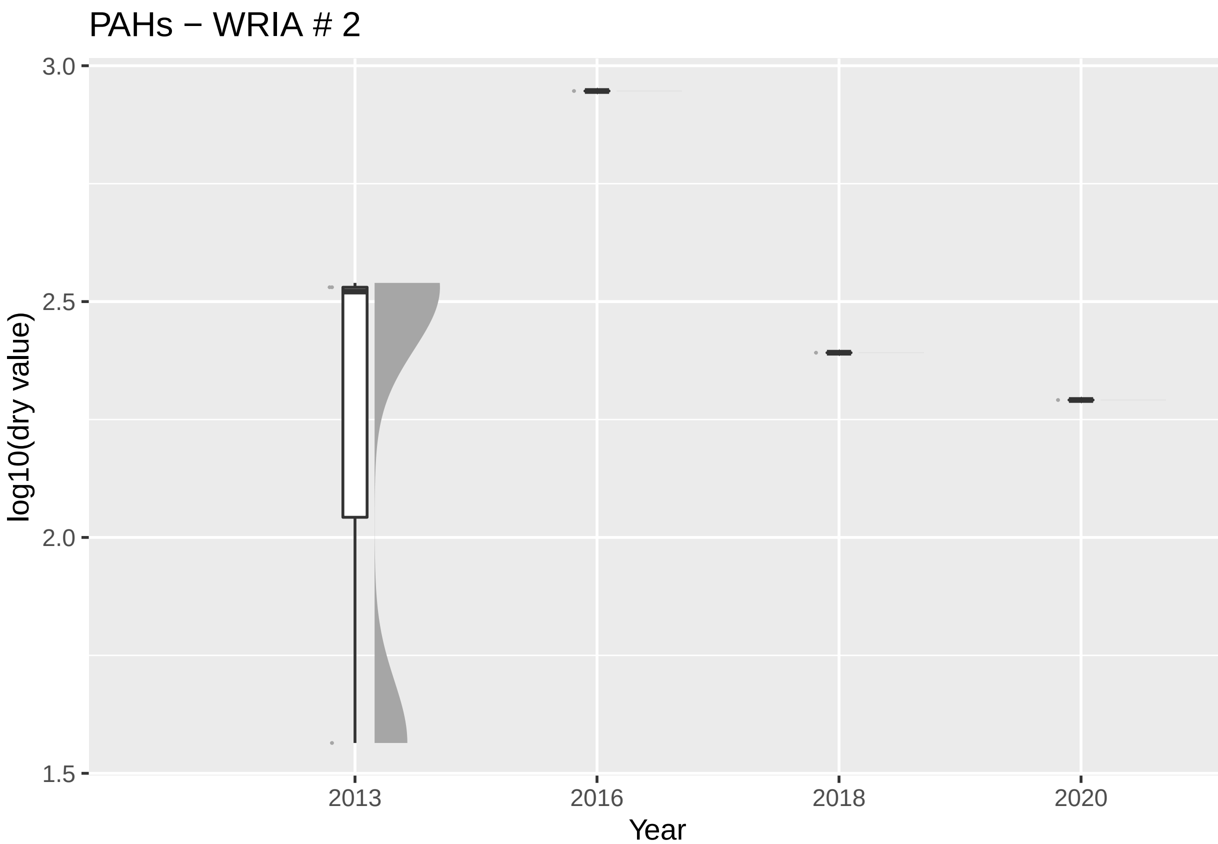
```
##
## $‘18‘
```




```
##
## $`3`
```


PAHs − WRIA # 3
log10(dry value)
3.0
2.5
2.0
1.5
2013
2016
2018
2020
Year

```
##
## $`2`
```

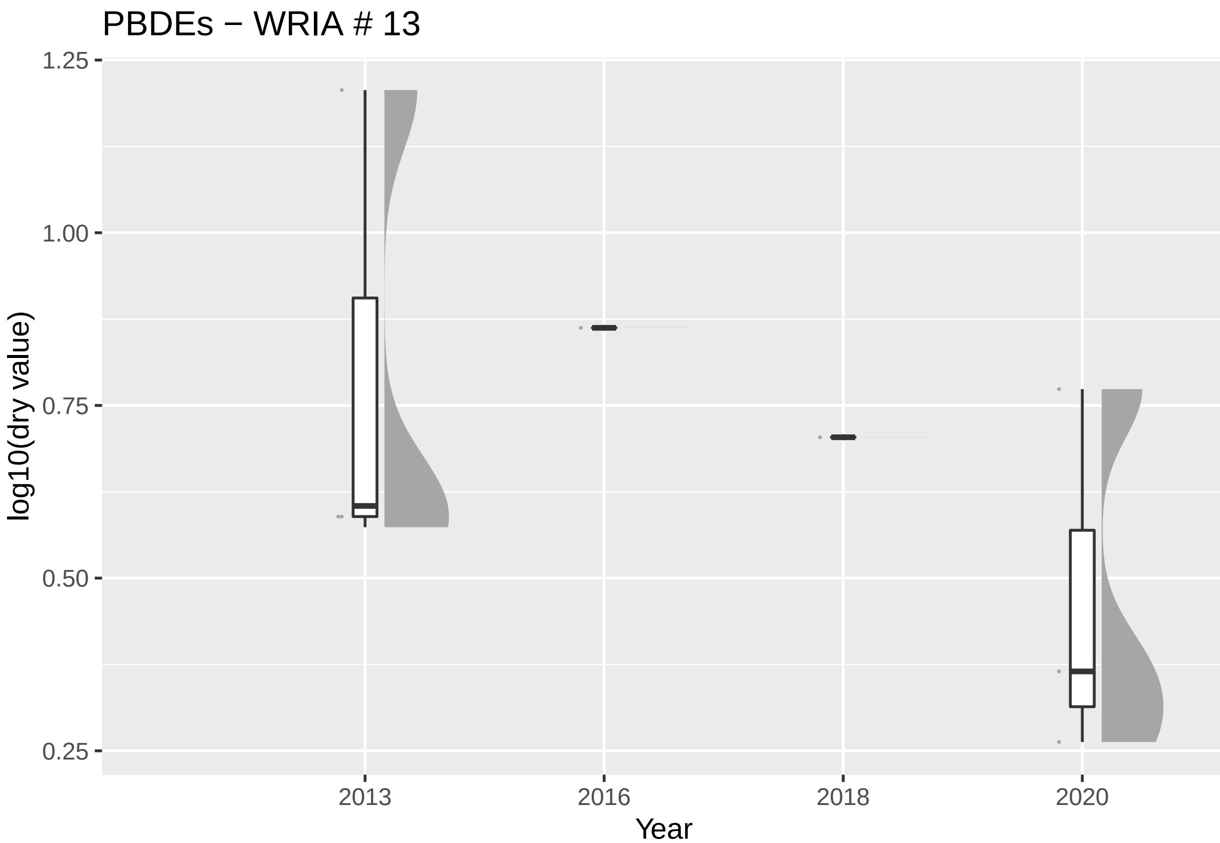

PAHs − WRIA # 2
3.0
2.5
2.0
1.5
log10(dry value)
2013
2016
2018
2020
Year

```
##
## $‘1‘
```

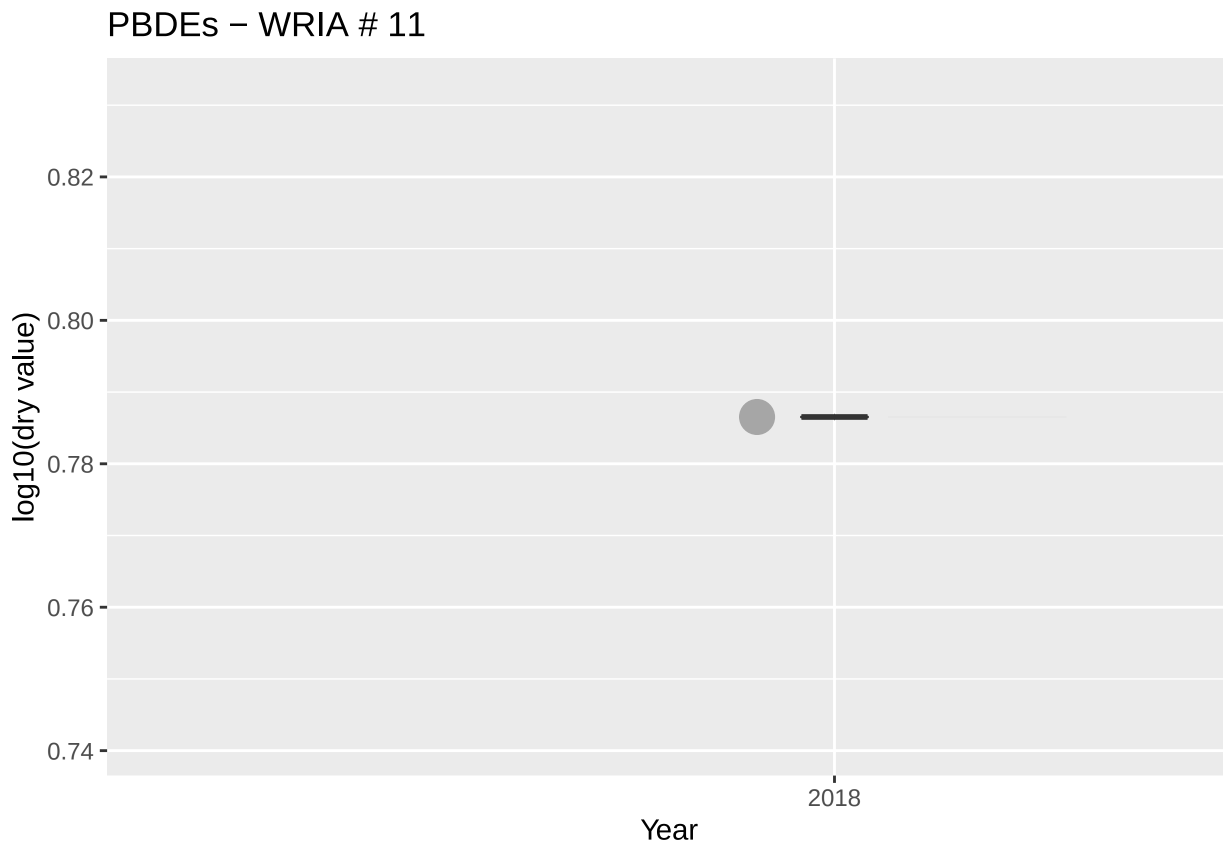

PAHs − WRIA # 1
log10(dry value)
2.6
2.4
2.2
2.0
1.8
2013
2016
2018
2020
Year

6.2.2 PBDE

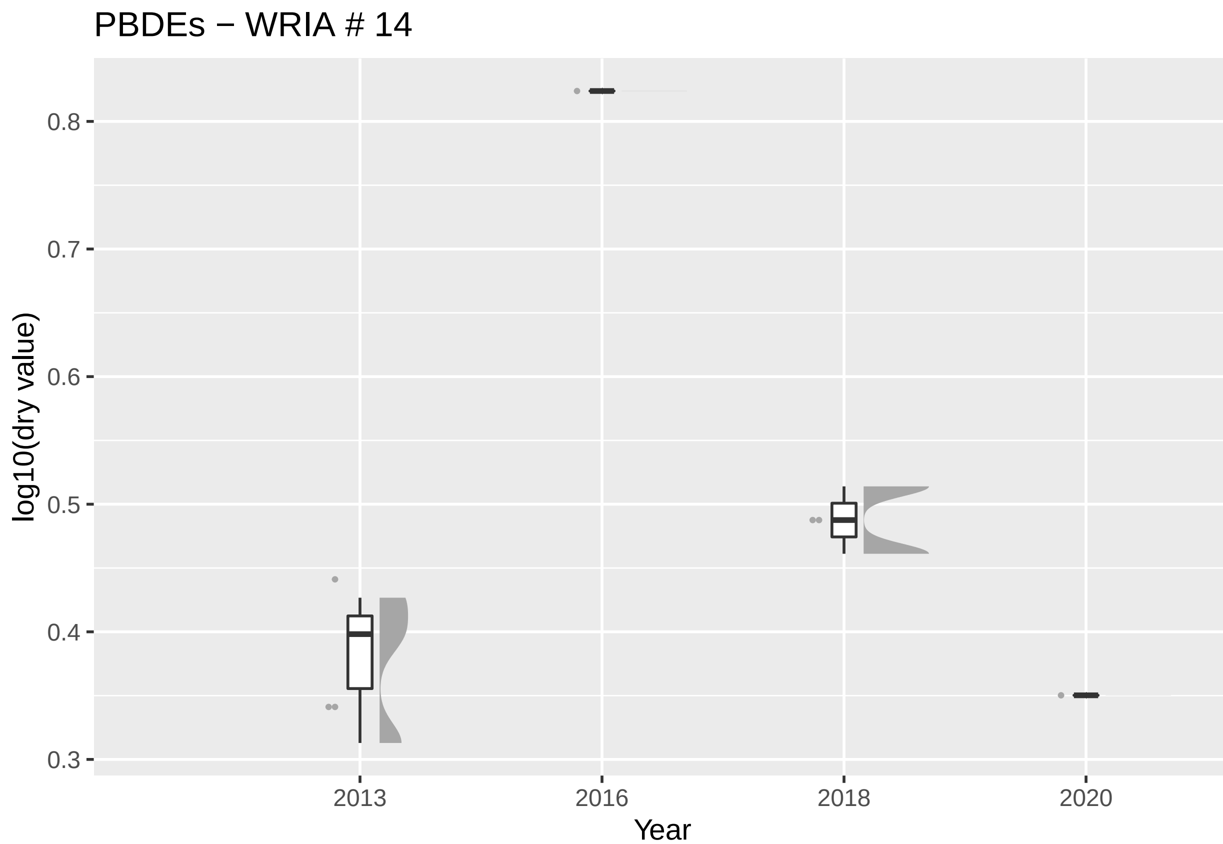
```
## $‘13‘
```



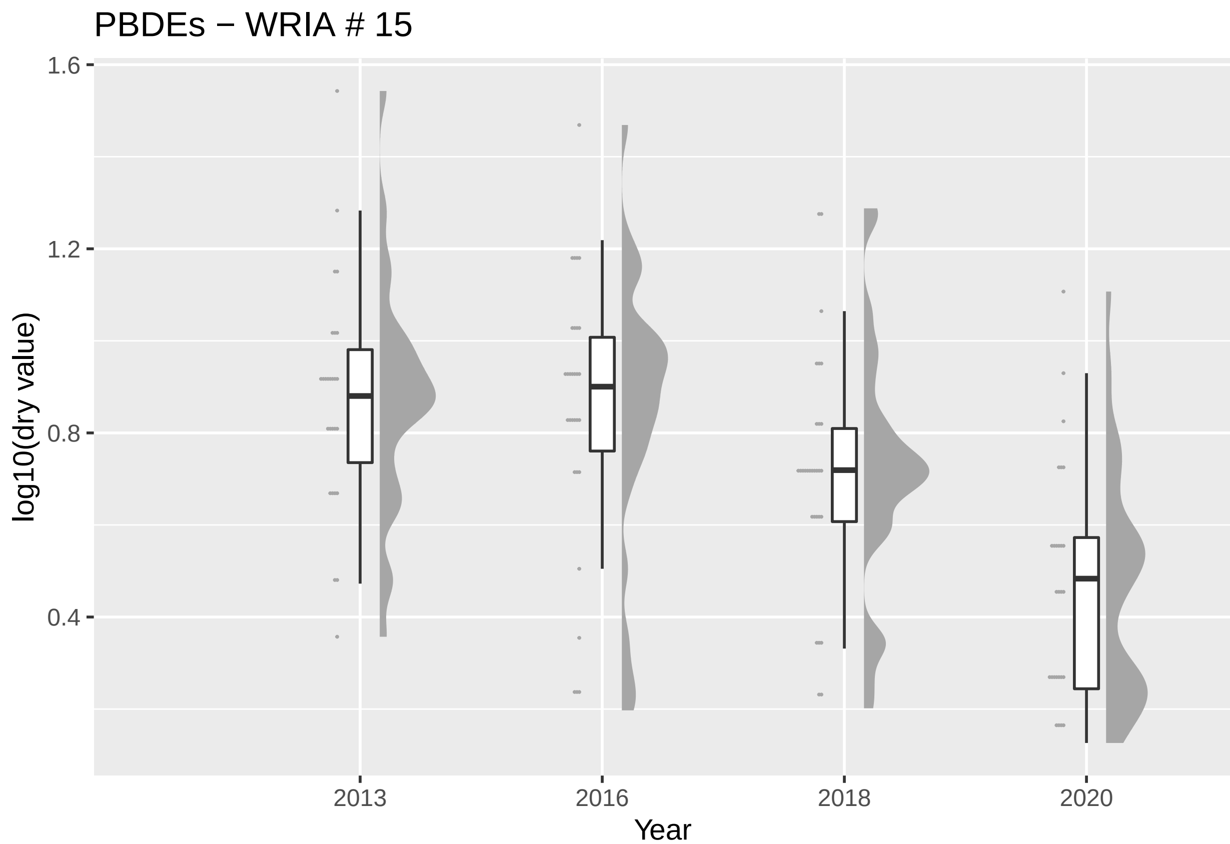

```
##
## $‘11‘
```



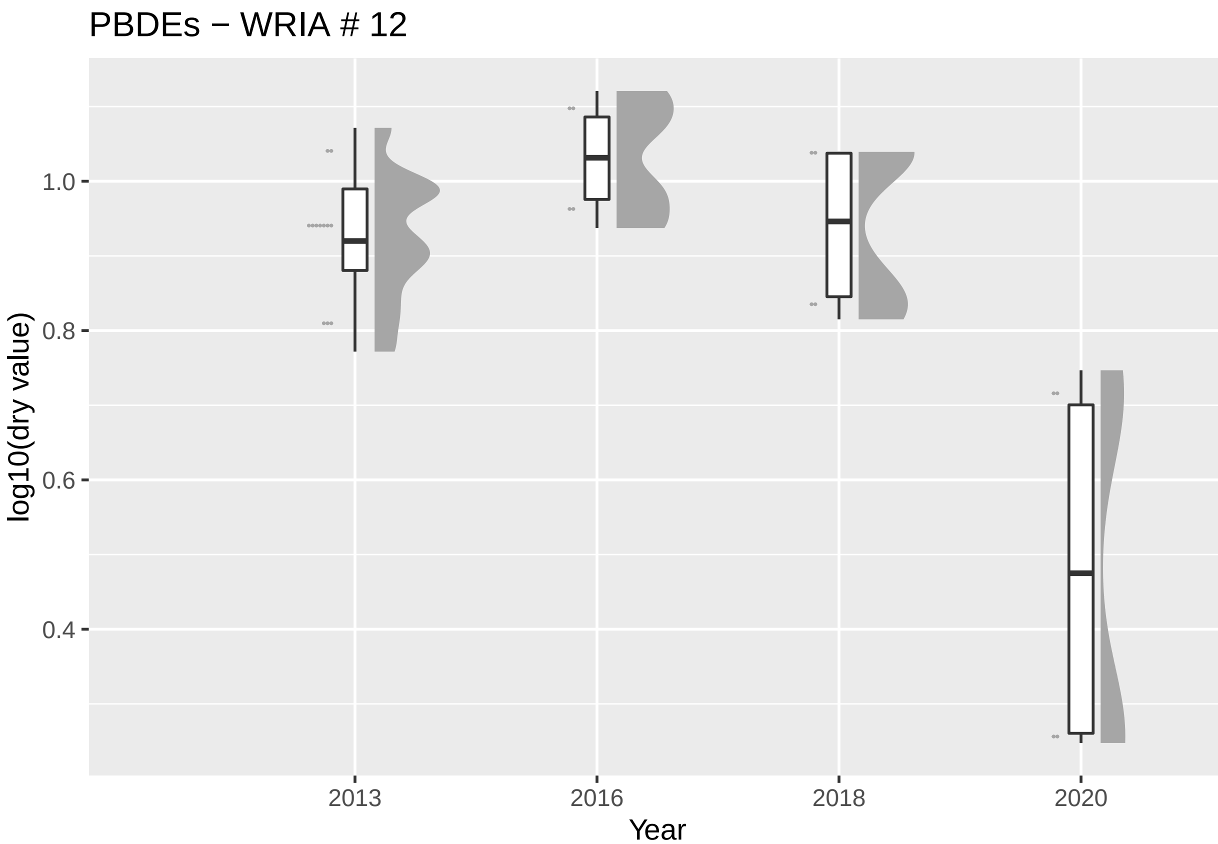

```
##
## $‘14‘
```


PBDEs − WRIA # 14
log10(dry value)
0.8
0.7
0.6
0.5
0.4
0.3
2013
2016
2018
2020
Year

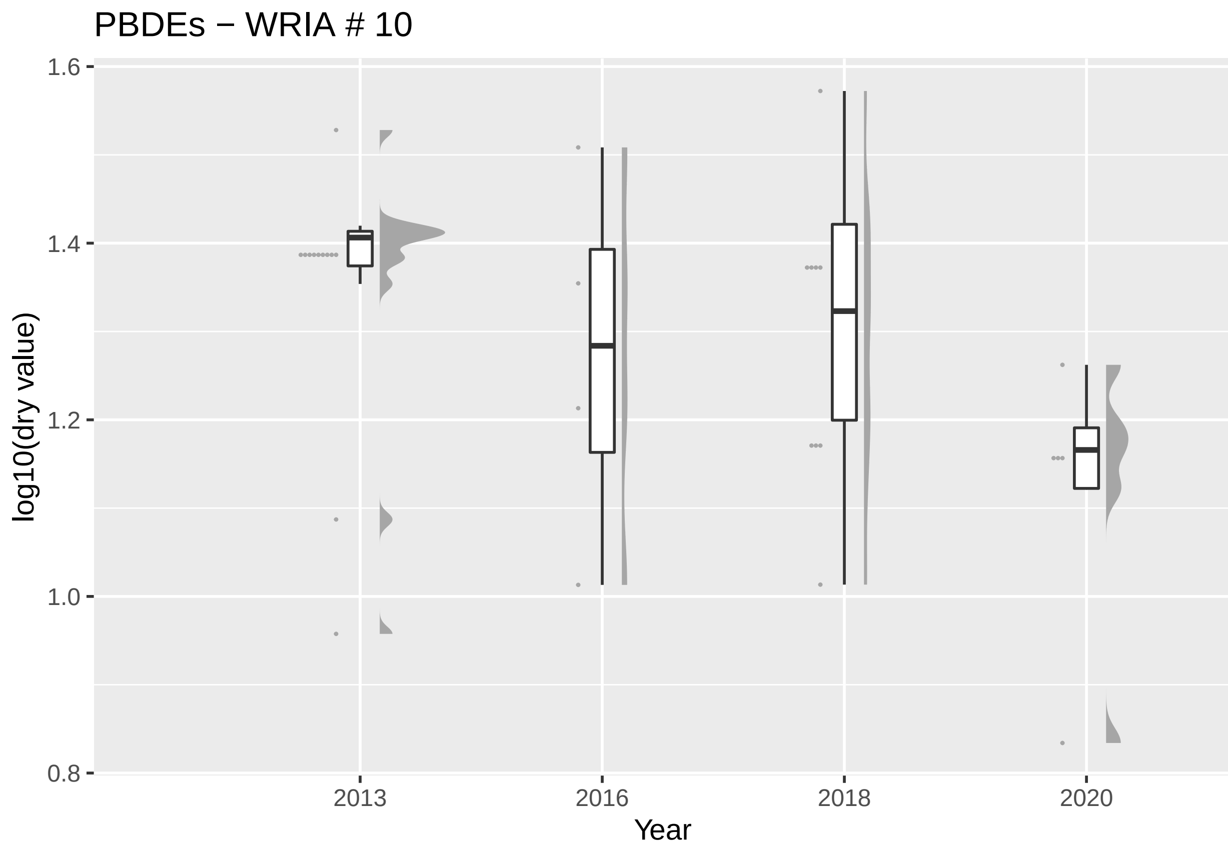
```
##
## $`15`
```


PBDEs − WRIA # 15
1.6
1.2
0.8
0.4
log10(dry value)
2013
2016
2018
2020
Year

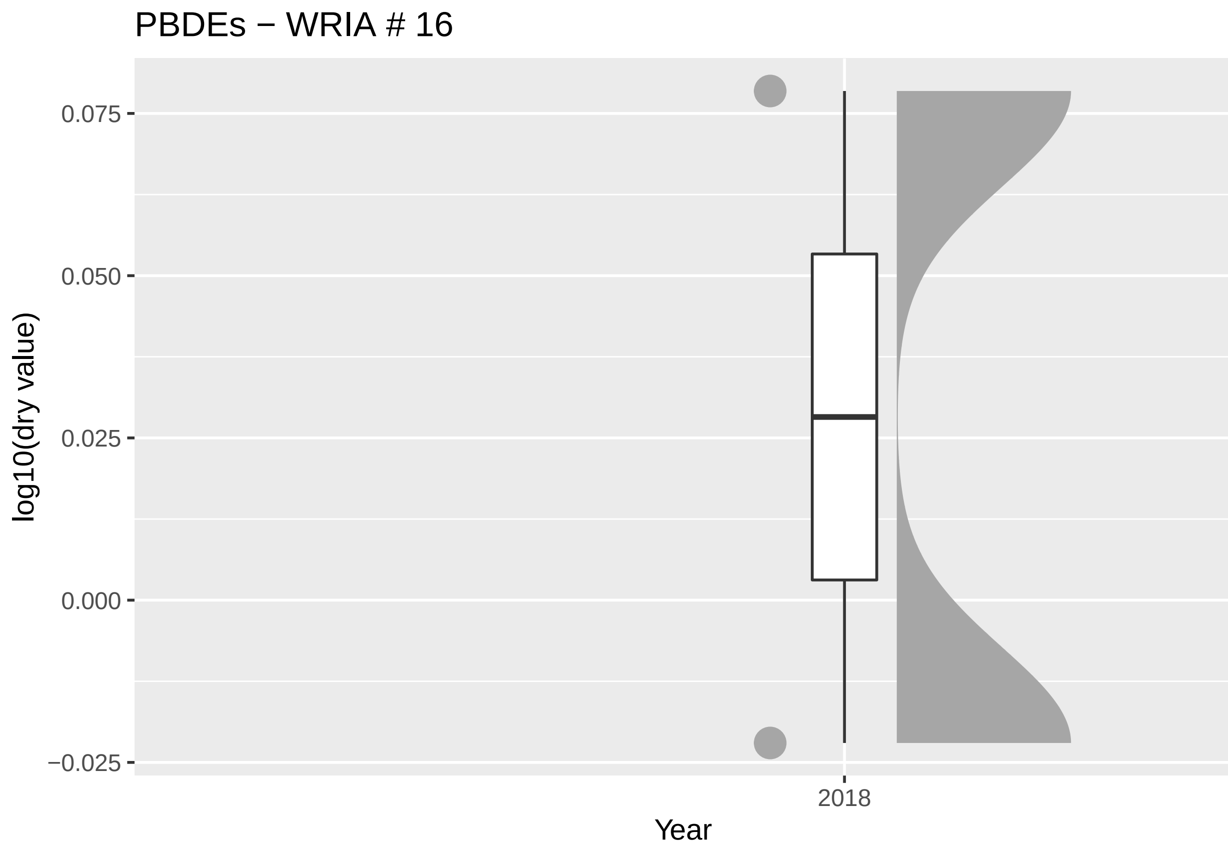
```
##
## $‘12‘
```


PBDEs − WRIA # 12
log10(dry value)
1.0
0.8
0.6
0.4
2013
2016
2018
2020
Year

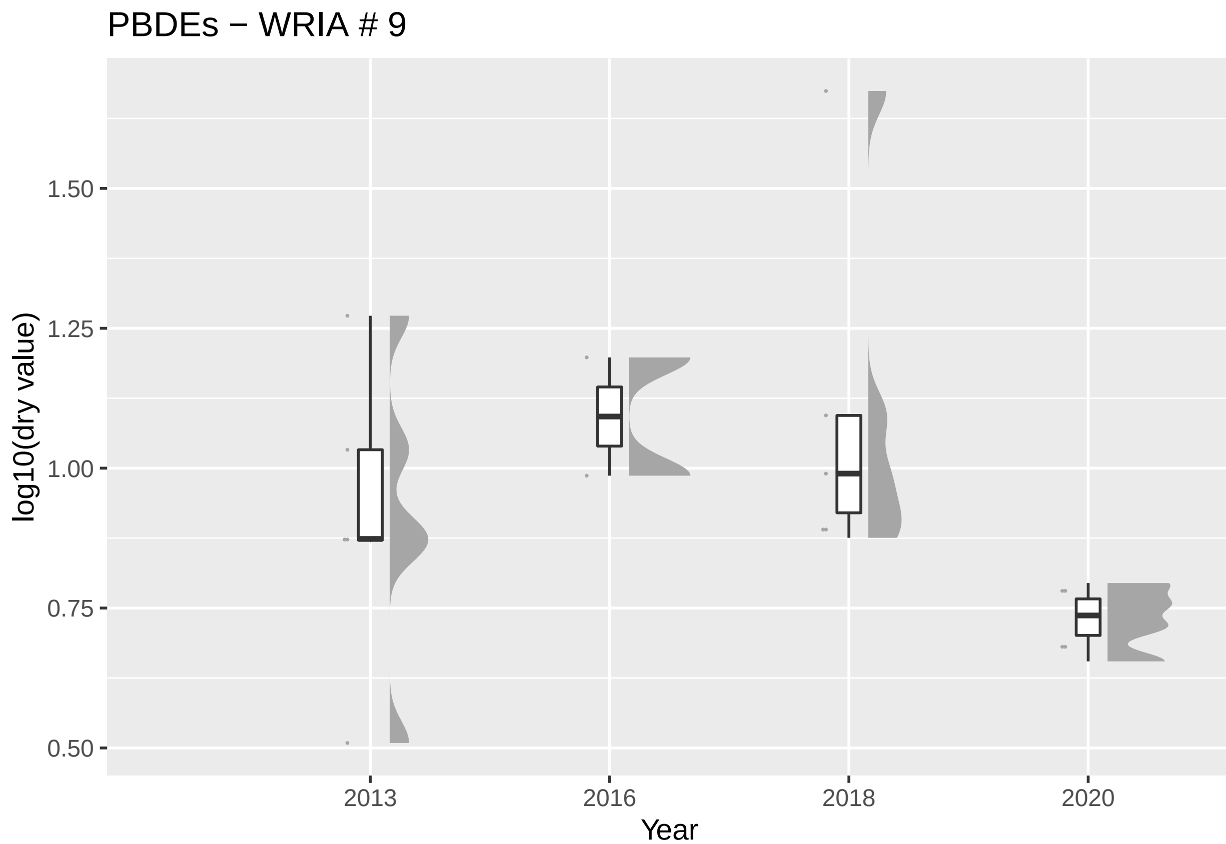
```
##
## $`10`
```


PBDEs − WRIA # 10
log10(dry value)
1.6
1.4
1.2
1.0
0.8
2013
2016
2018
2020
Year

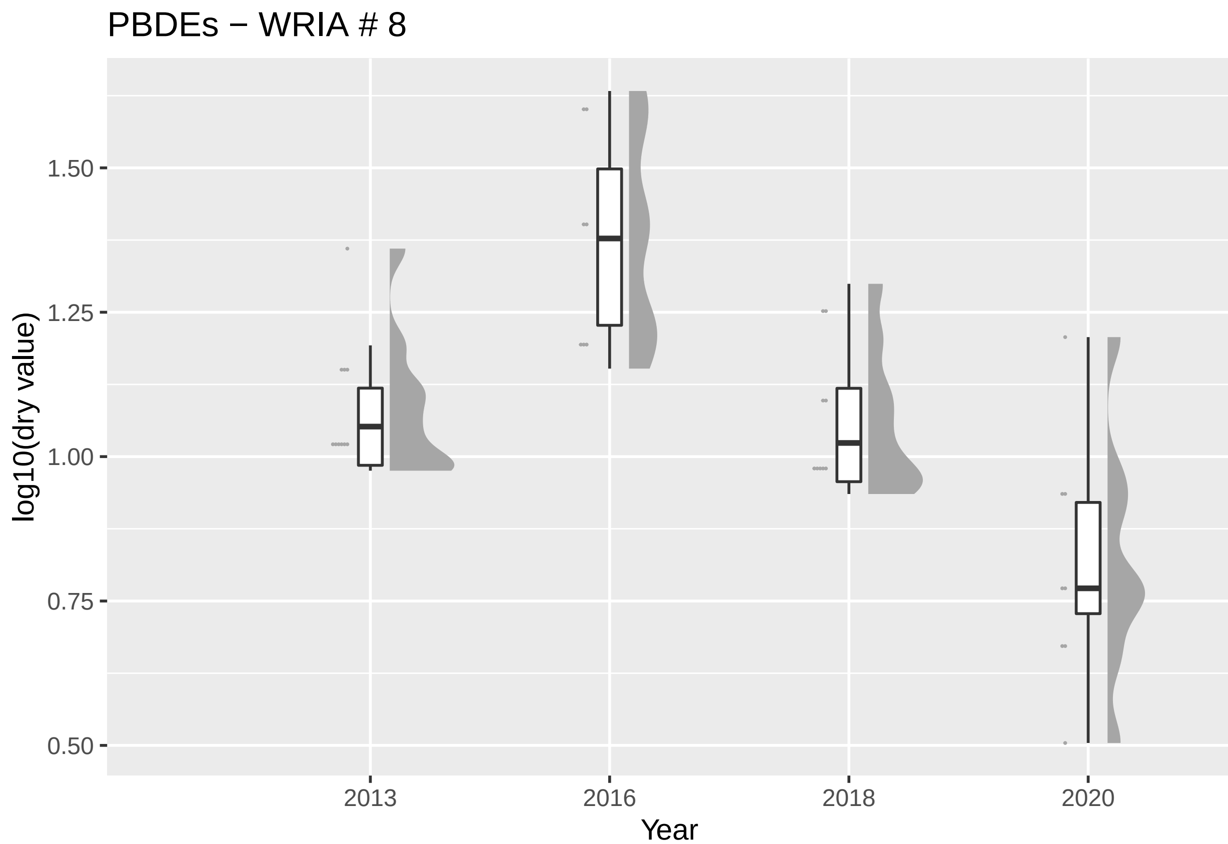
```
##
## $`16`
```


PBDEs − WRIA # 16
0.075
0.050
0.025
0.000
−0.025
log10(dry value)
2018
Year

```
##
## $`9`
```


PBDEs − WRIA # 9
log10(dry value)
1.50
1.25
1.00
0.75
0.50
2013
2016
2018
2020
Year

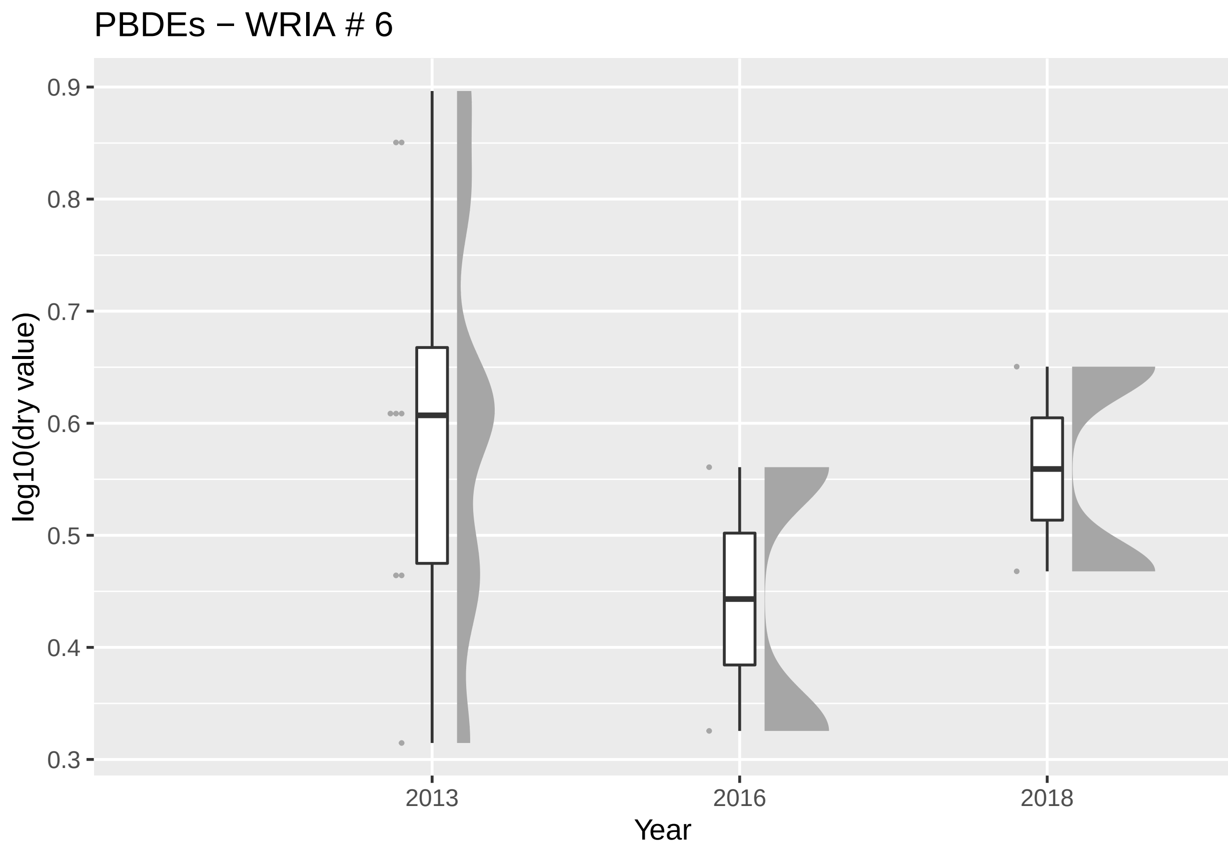
```
##
## $`8`
```


PBDEs − WRIA # 8
log10(dry value)
1.50
1.25
1.00
0.75
0.50
2013
2016
2018
2020
Year

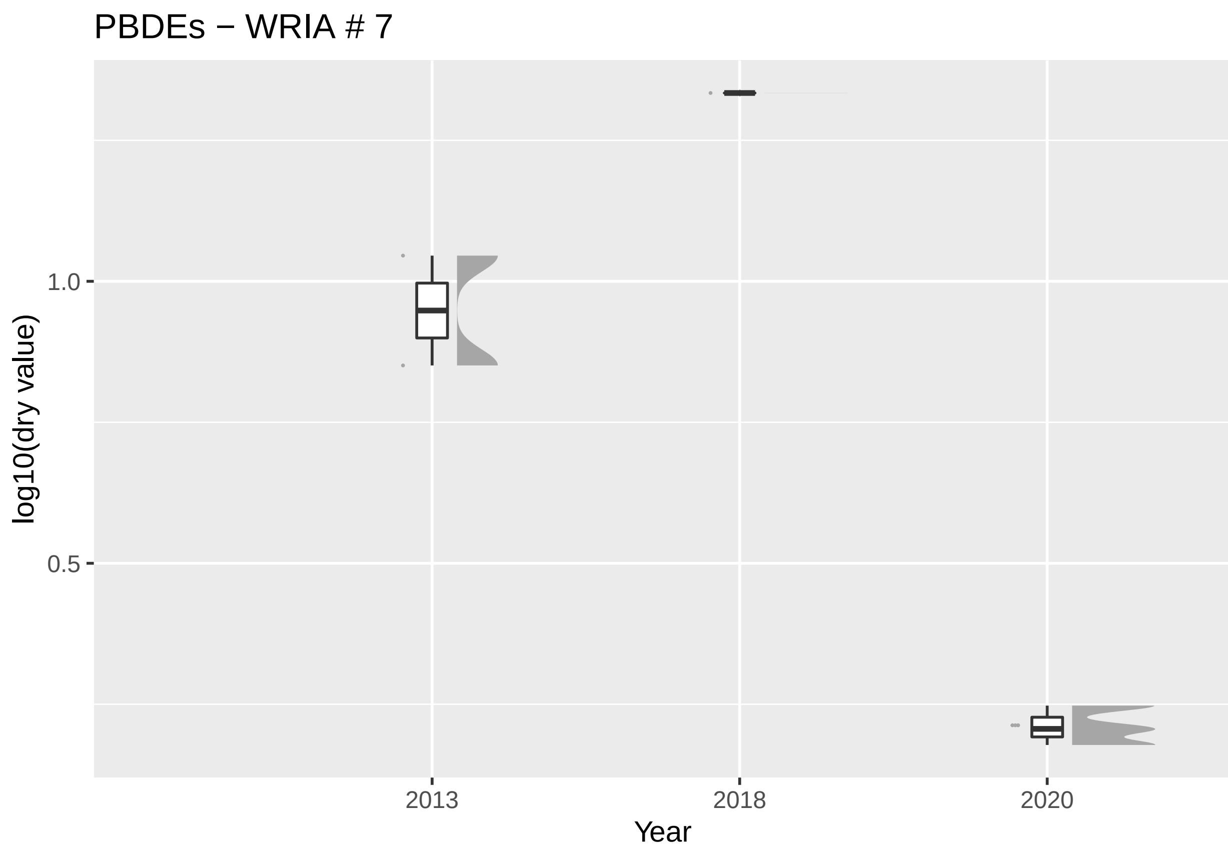
```
##
## $‘17‘
```


PBDEs − WRIA # 17
log10(dry value)
1.2
0.8
0.4
0.0
2013
2016
2018
2020
Year

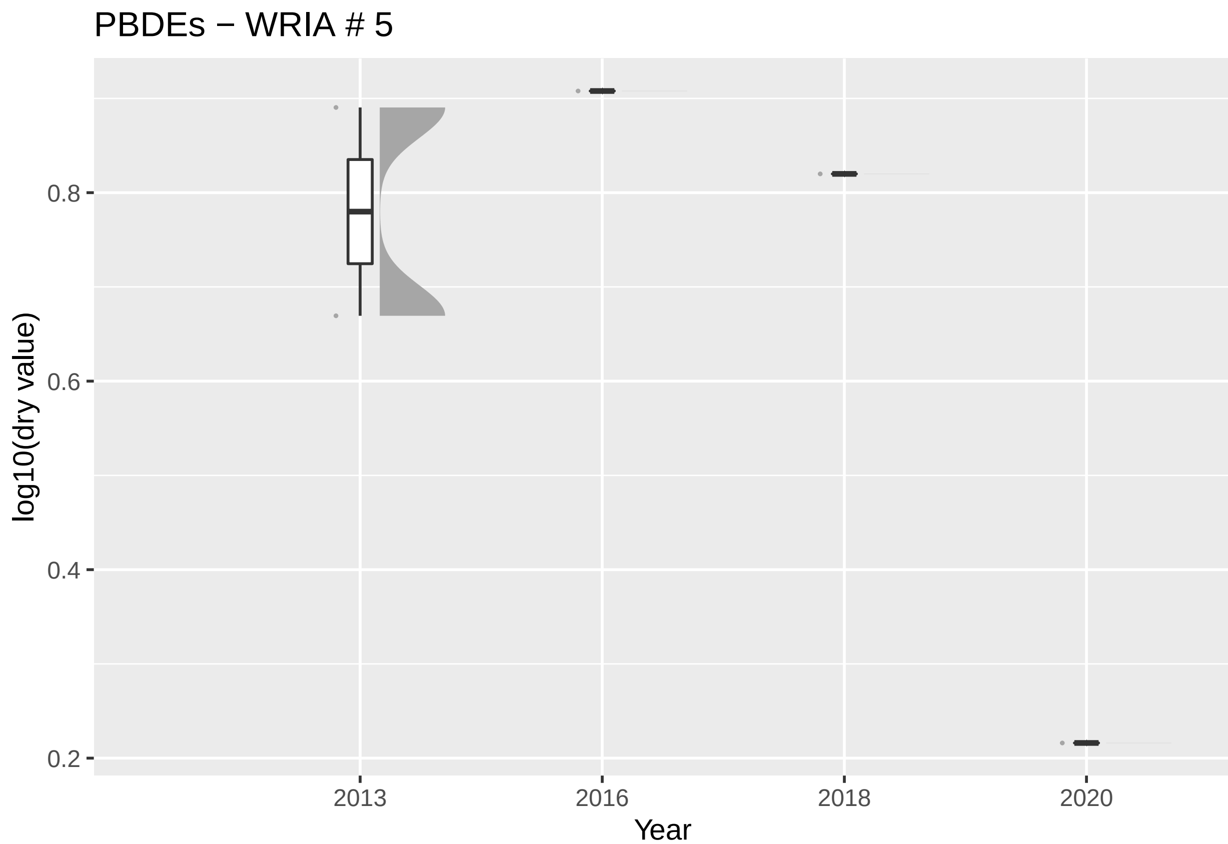
```
##
## $‘6‘
```


PBDEs − WRIA # 6
log10(dry value)
0.9
0.8
0.7
0.6
0.5
0.4
0.3
2013
2016
2018
Year

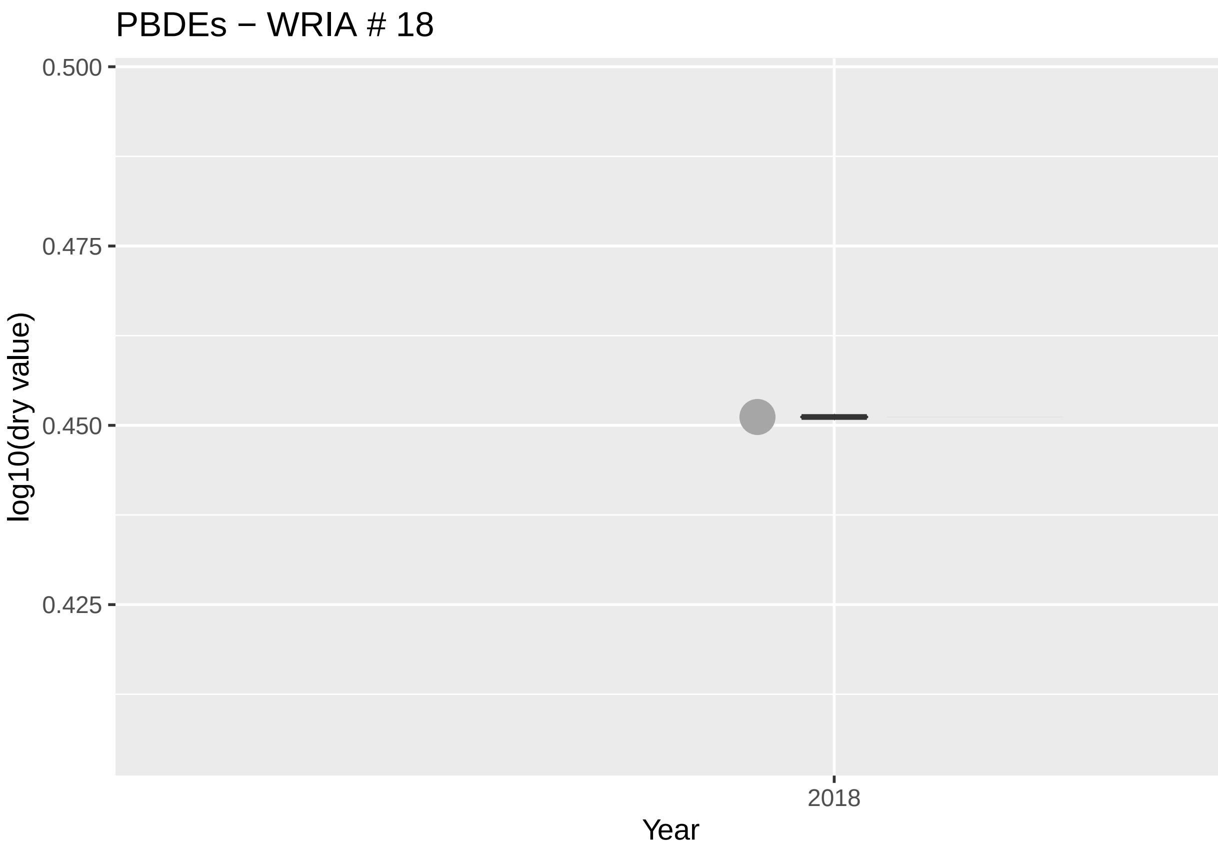
```
##
## $‘7‘
```



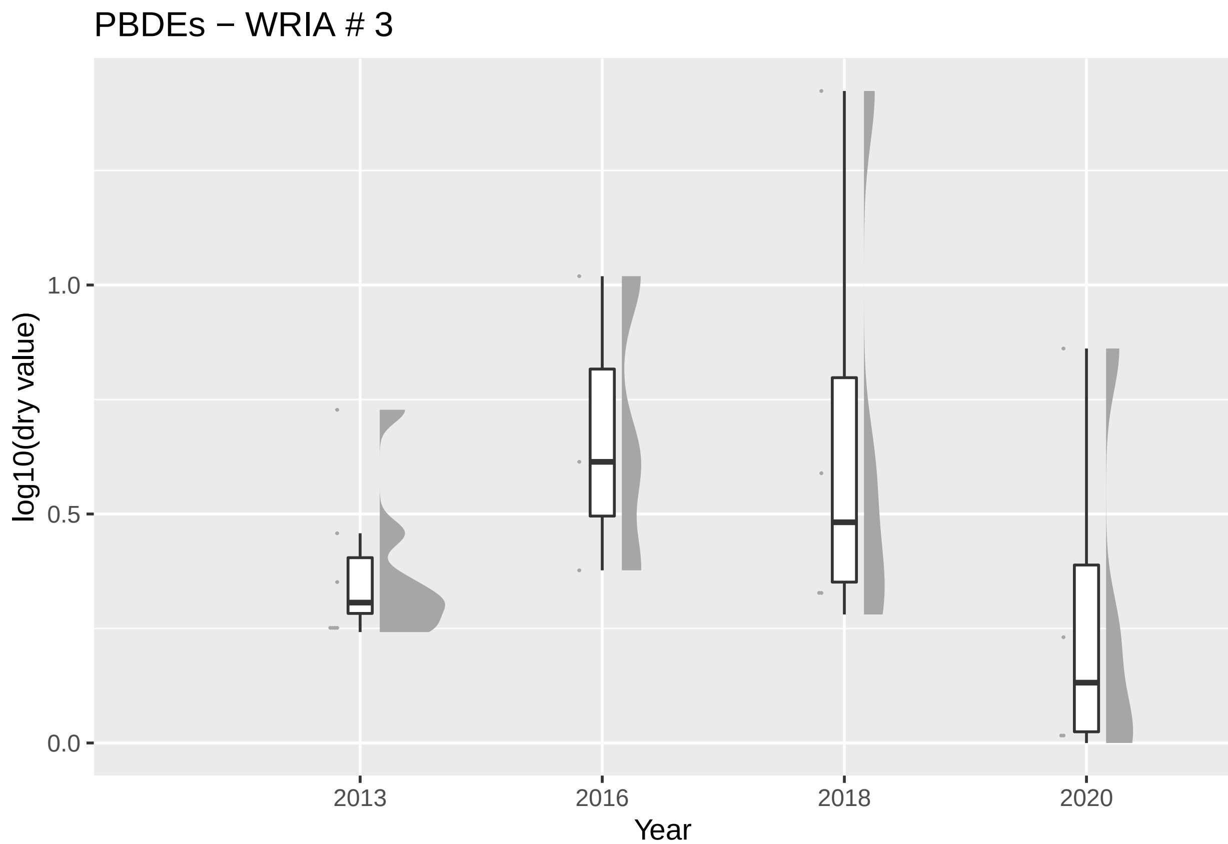

```
##
## $‘5‘
```


PBDEs − WRIA # 5
log10(dry value)
0.8
0.6
0.4
0.2
2013
2016
2018
2020
Year

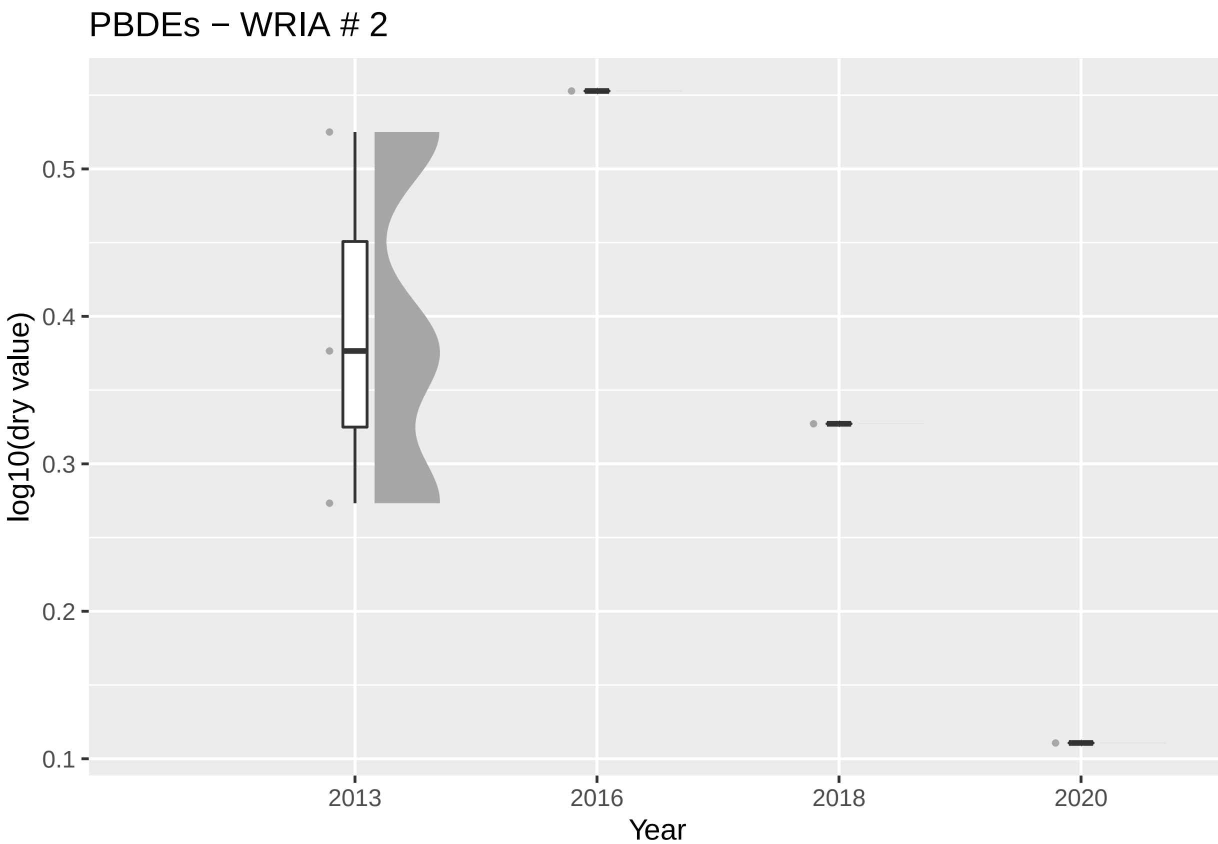
```
##
## $‘18‘
```



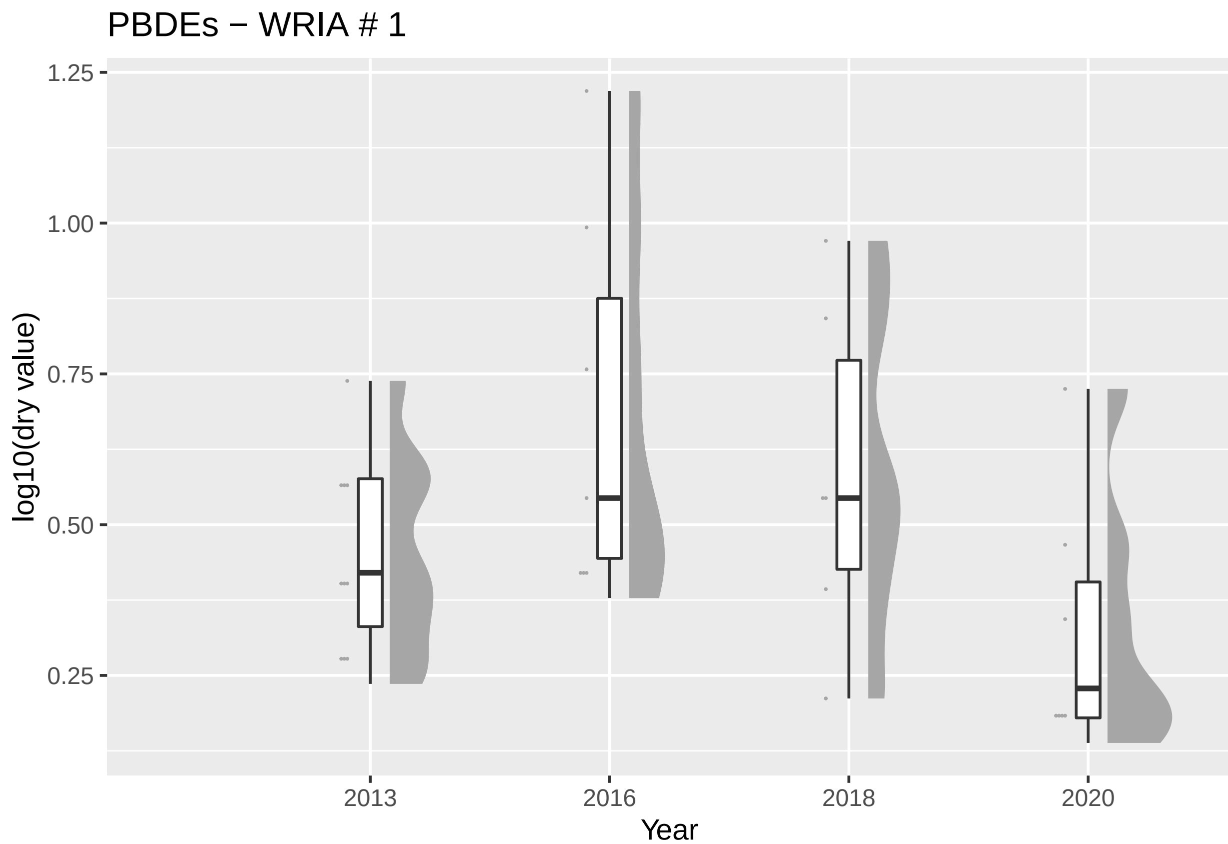

```
##
## $‘3‘
```




```
##
## $‘2‘
```


PBDEs − WRIA # 2
log10(dry value)
0.5
0.4
0.3
0.2
0.1
2013
2016
2018
2020
Year

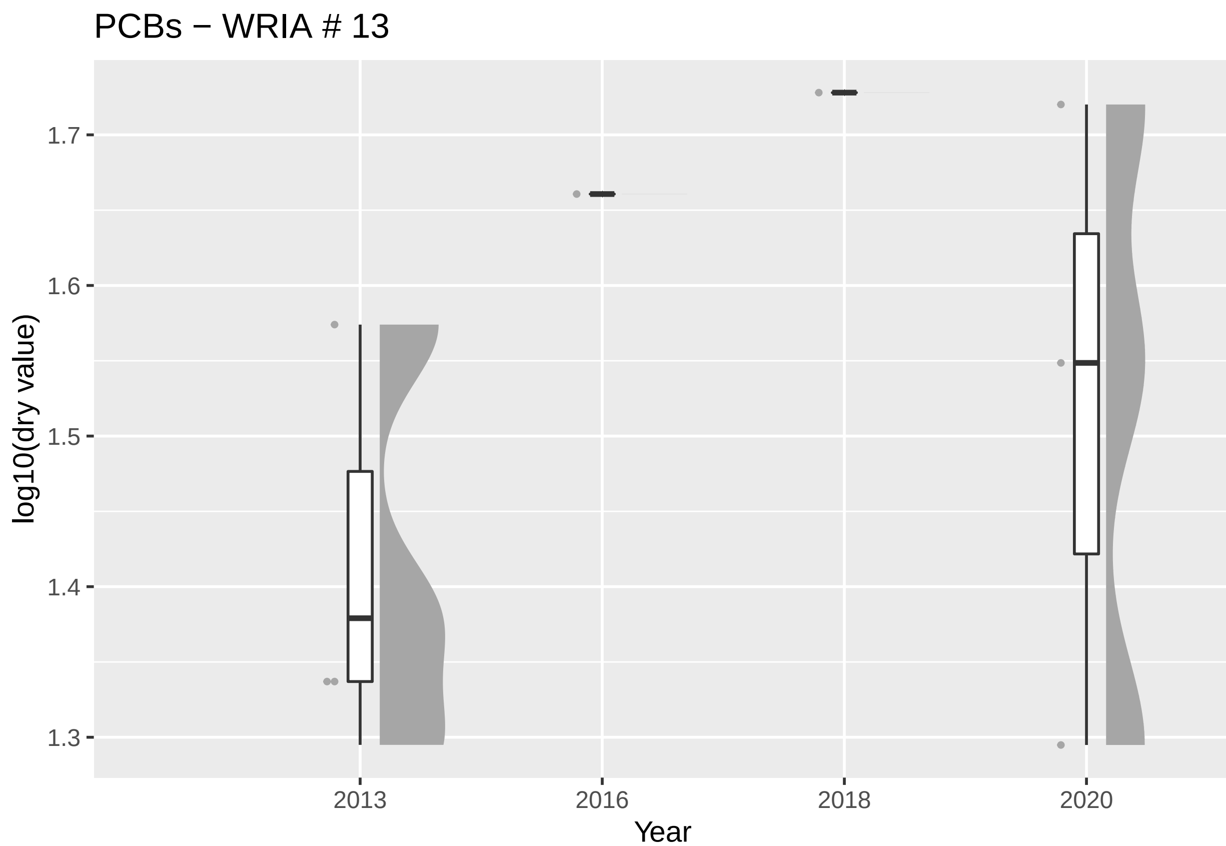
```
##
## $‘1‘
```


PBDEs − WRIA # 1
log10(dry value)
1.25
1.00
0.75
0.50
0.25
2013
2016
2018
2020
Year

277

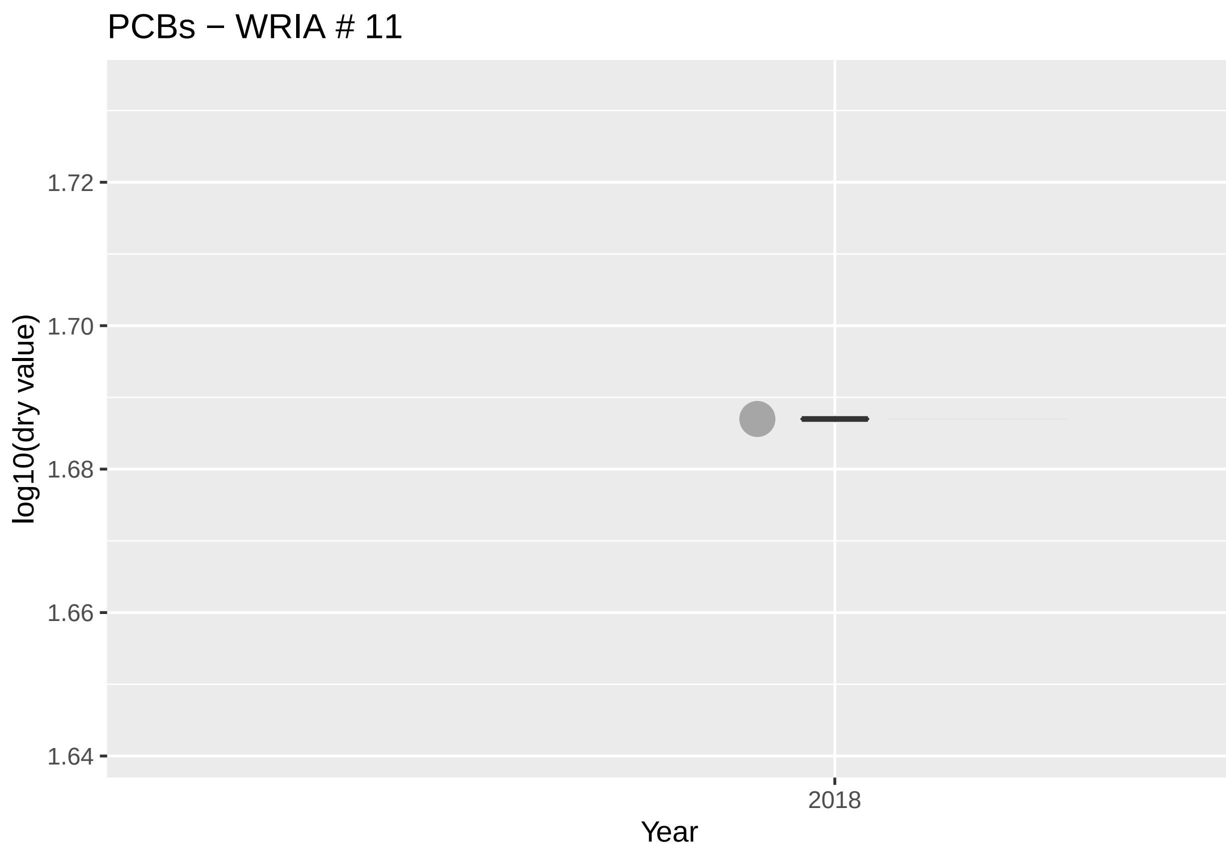
6.2.3 PCB

```
## $‘13‘
```

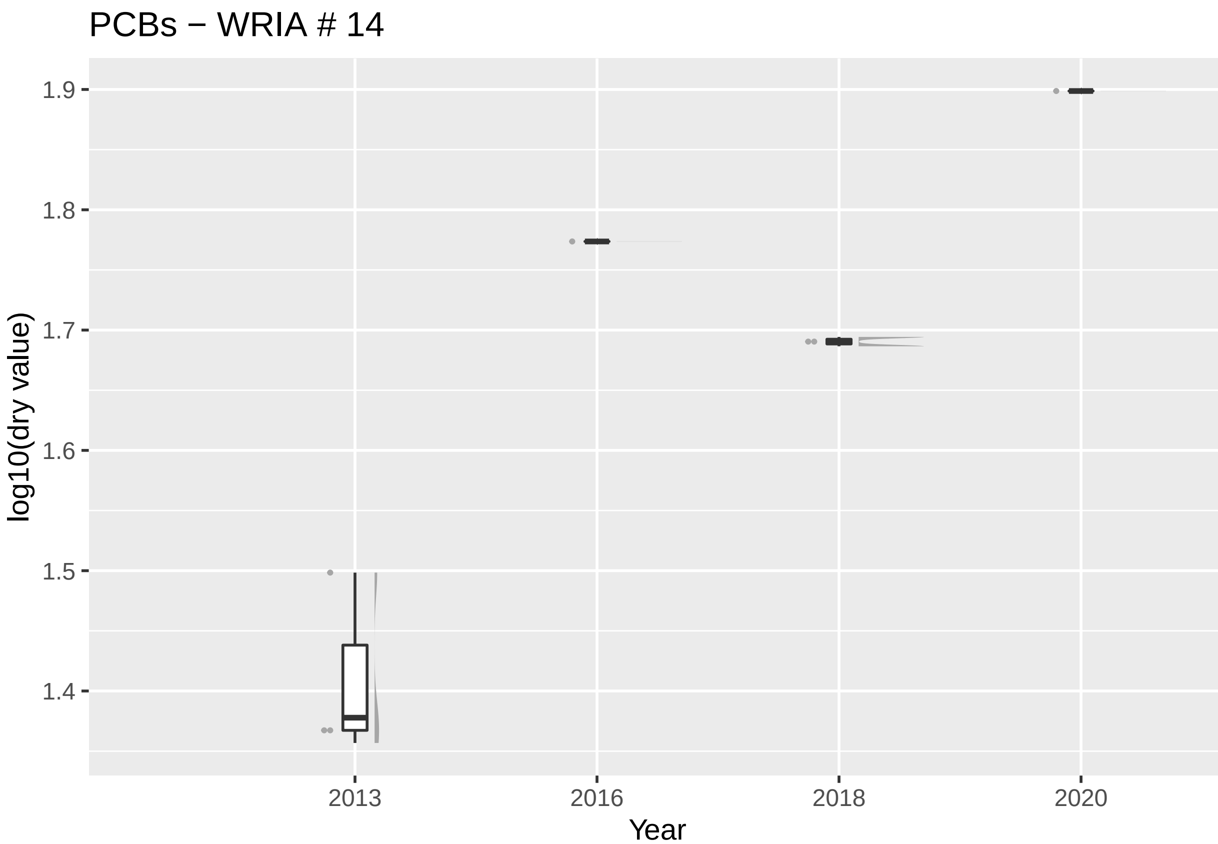


PCBs − WRIA # 13
log10(dry value)
1.7
1.6
1.5
1.4
1.3
2013
2016
2018
2020
Year

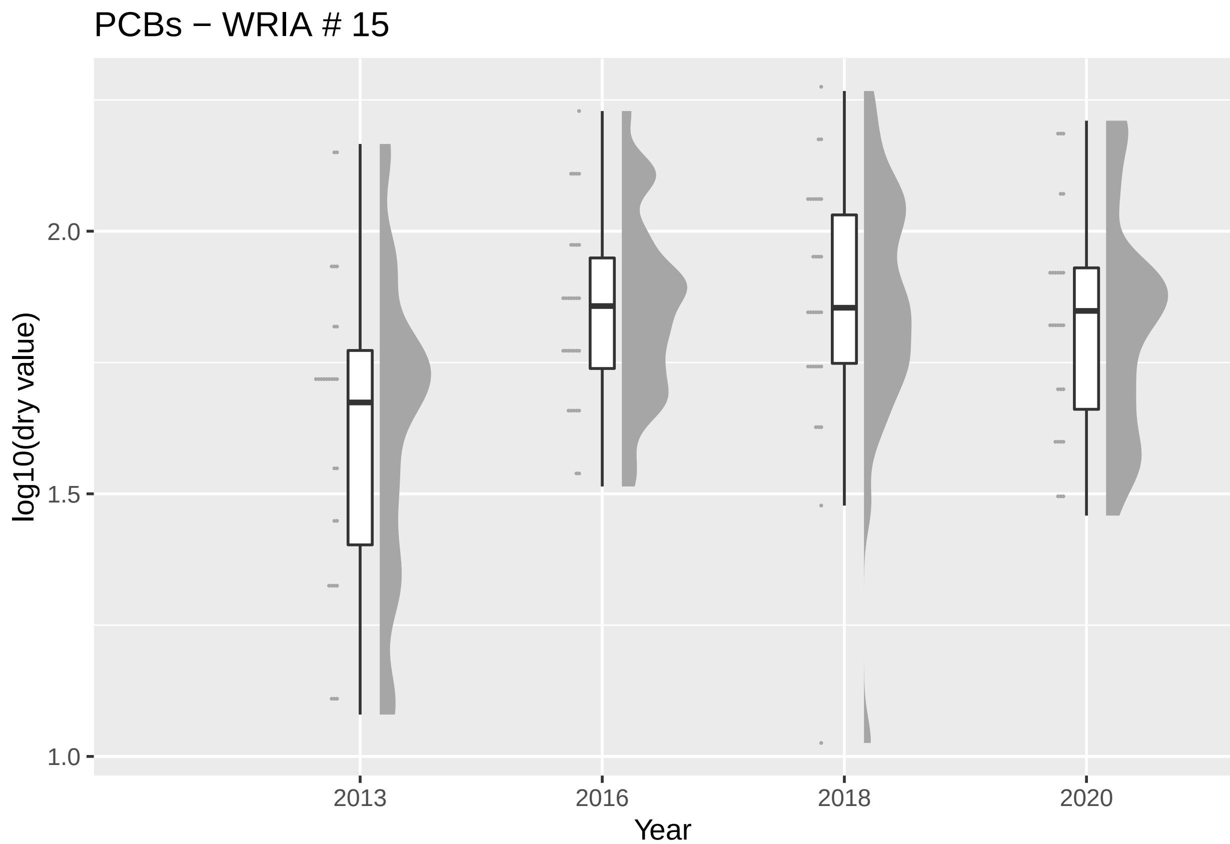

```
##
## $‘11‘
```



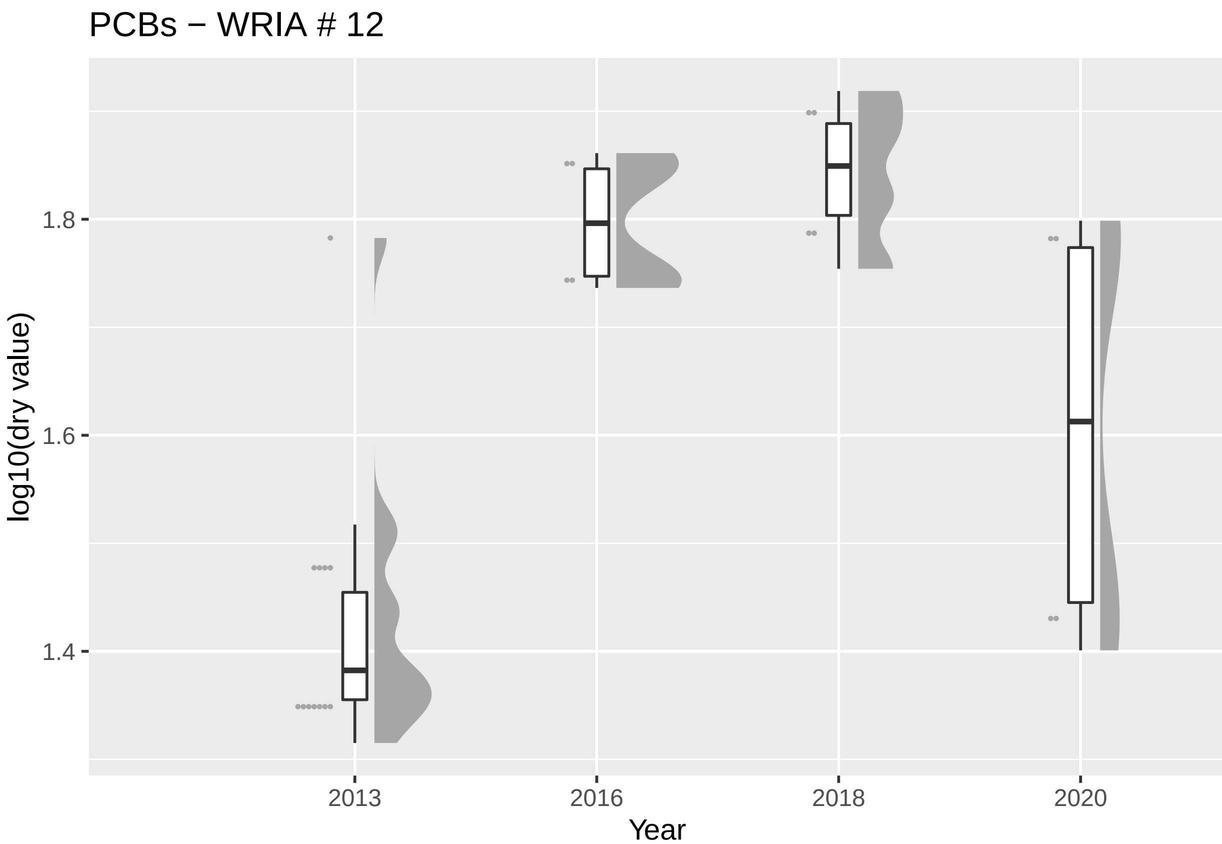

```
##
## $‘14‘
```


PCBs − WRIA # 14
log10(dry value)
1.9
1.8
1.7
1.6
1.5
1.4
2013
2016
2018
2020
Year

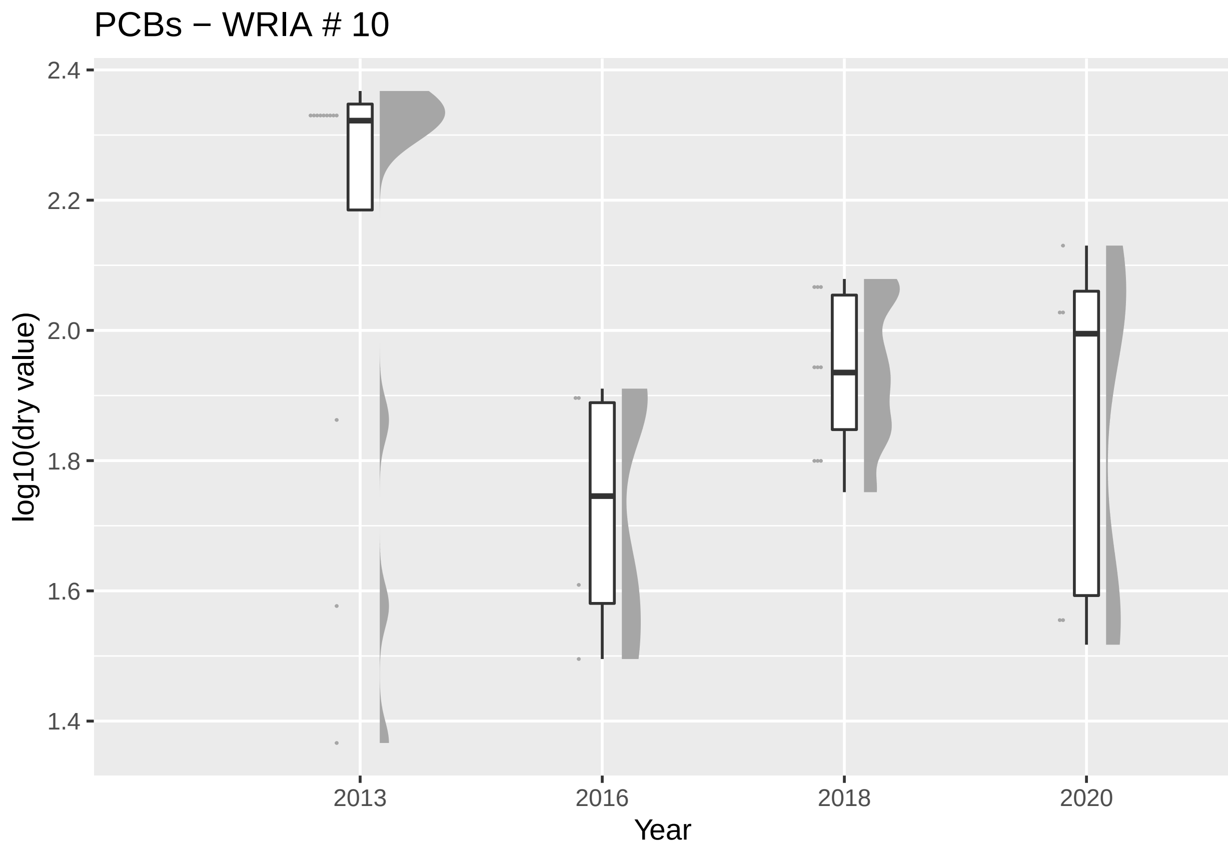
```
##
## $‘15‘
```



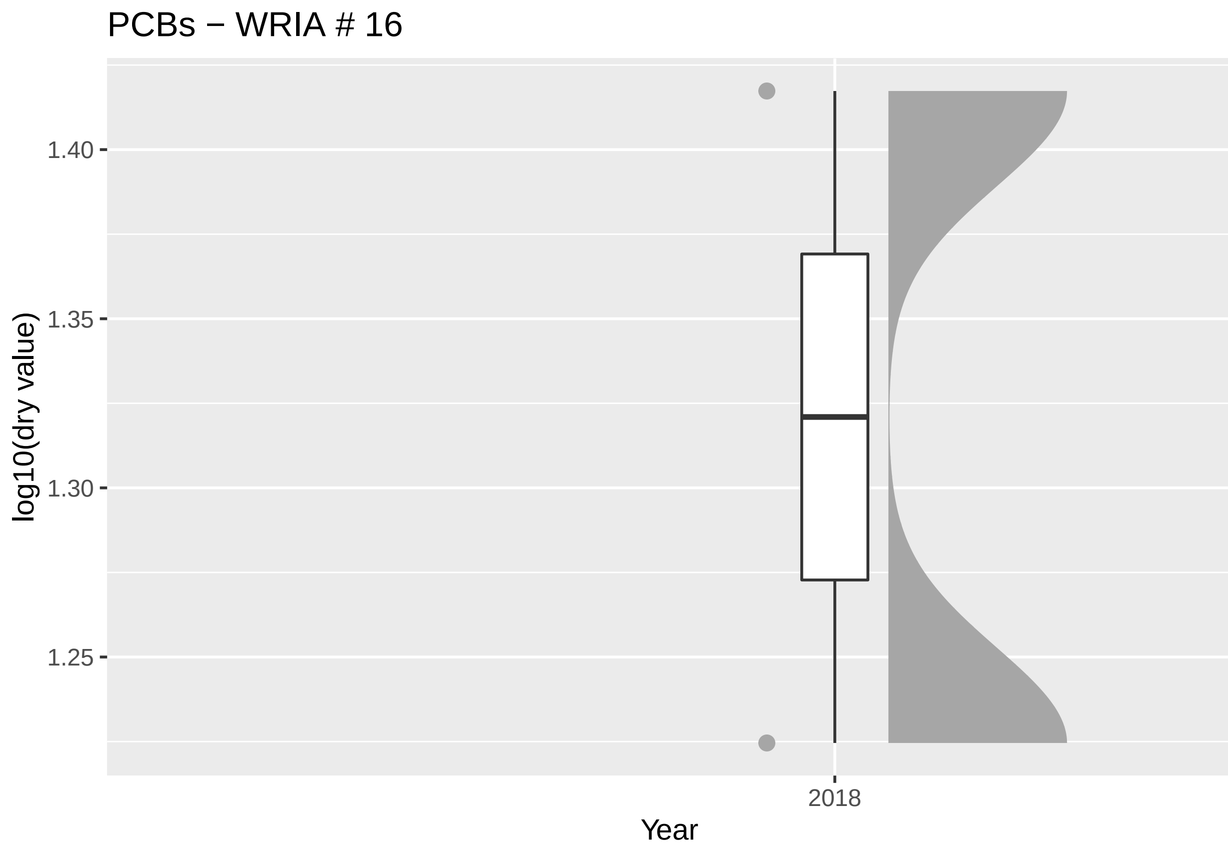

```
##
## $`12`
```



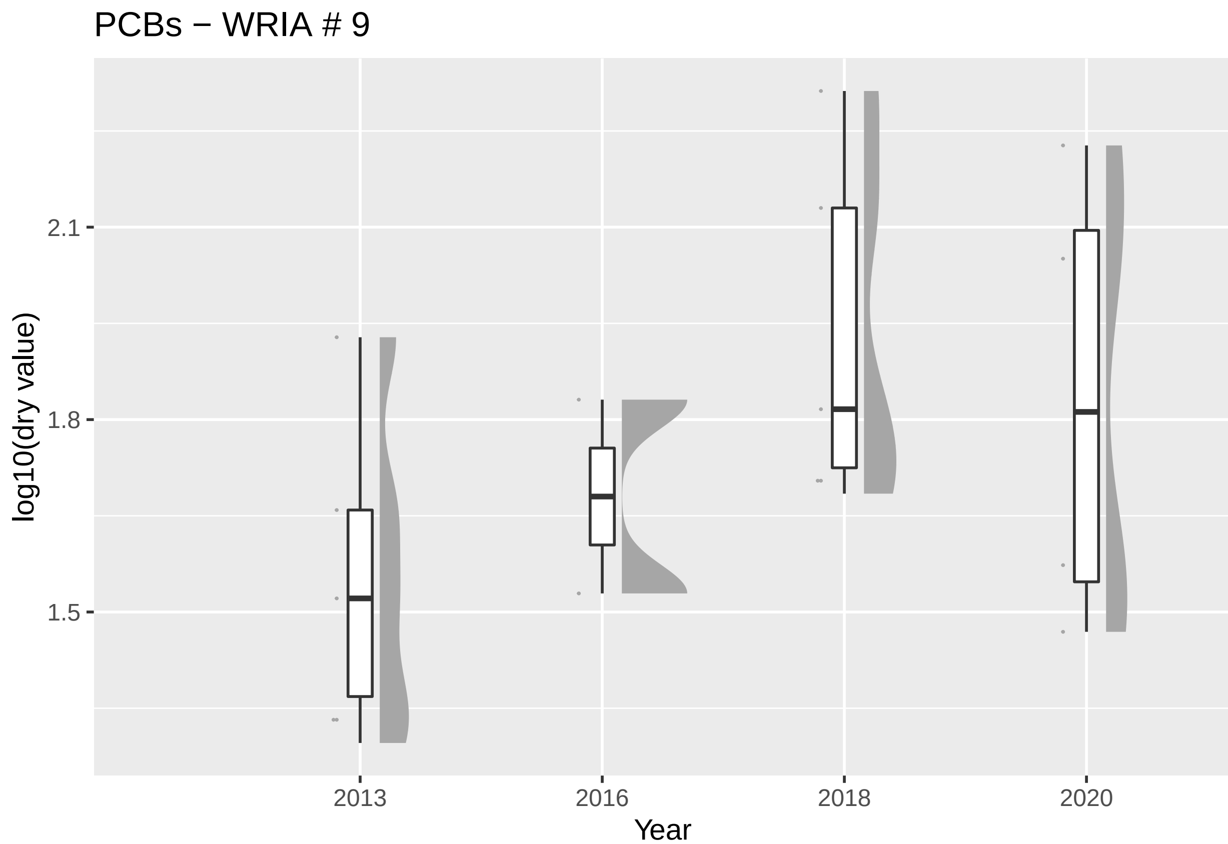

```
##
## $‘10‘
```


PCBs − WRIA # 10
log10(dry value)
2.4
2.2
2.0
1.8
1.6
1.4
2013
2016
2018
2020
Year

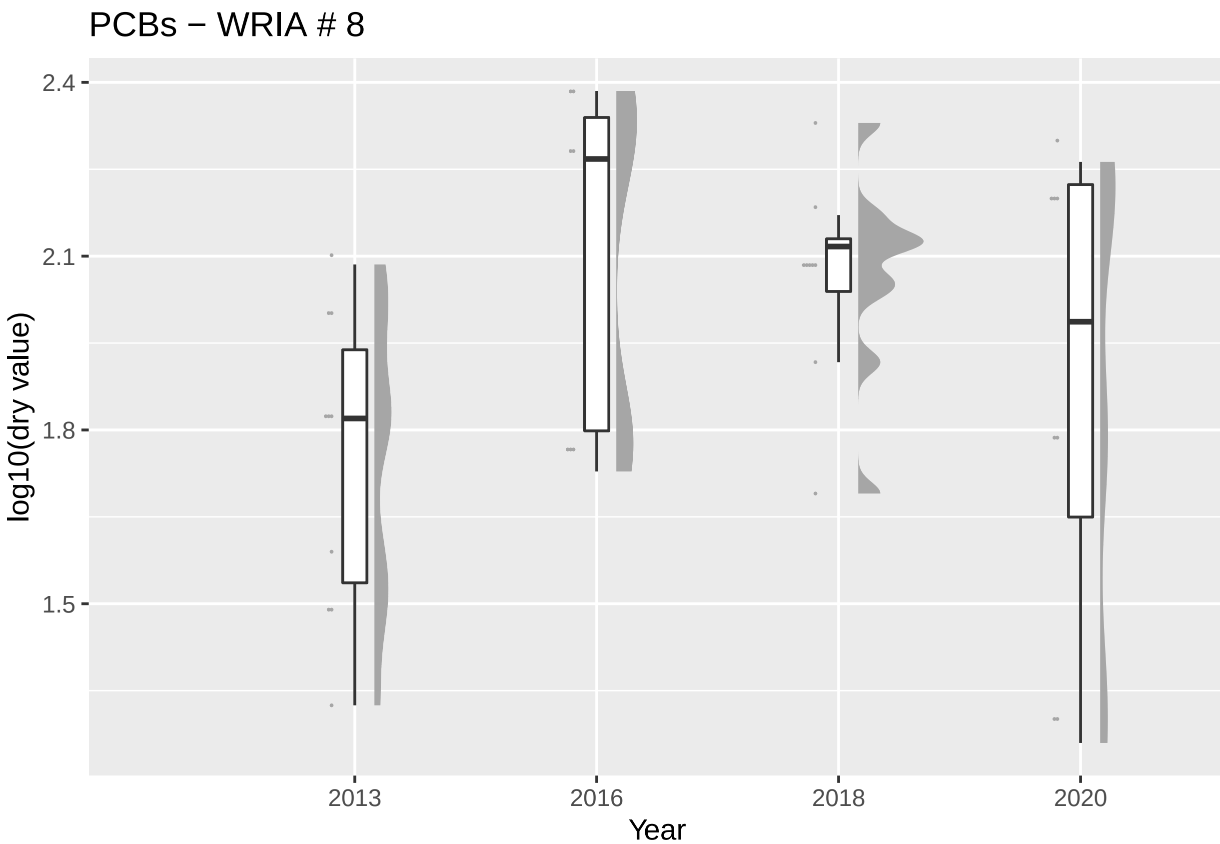
```
##
## $‘16‘
```




```
##
## $`9`
```



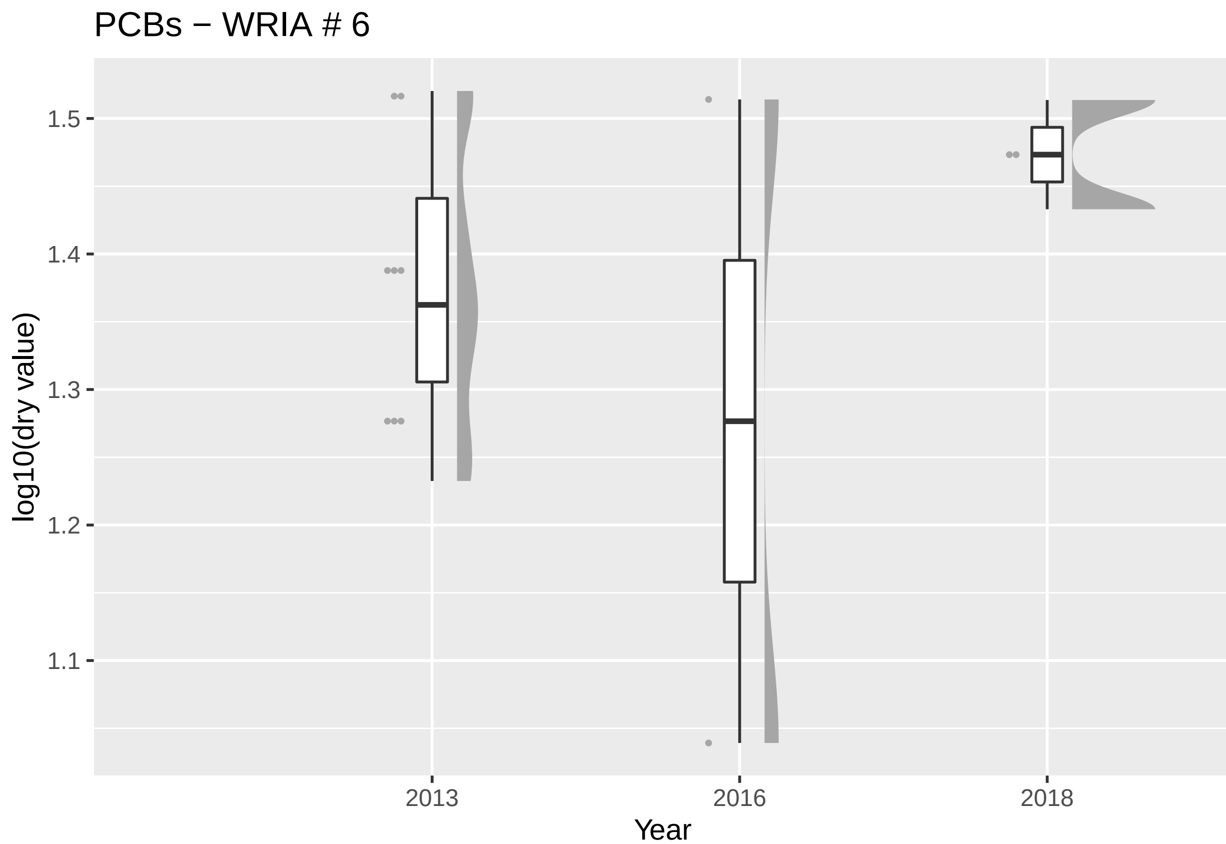

```
##
## $‘8‘
```


PCBs − WRIA # 8
2.4
2.1
1.8
1.5
log10(dry value)
2013
2016
2018
2020
Year

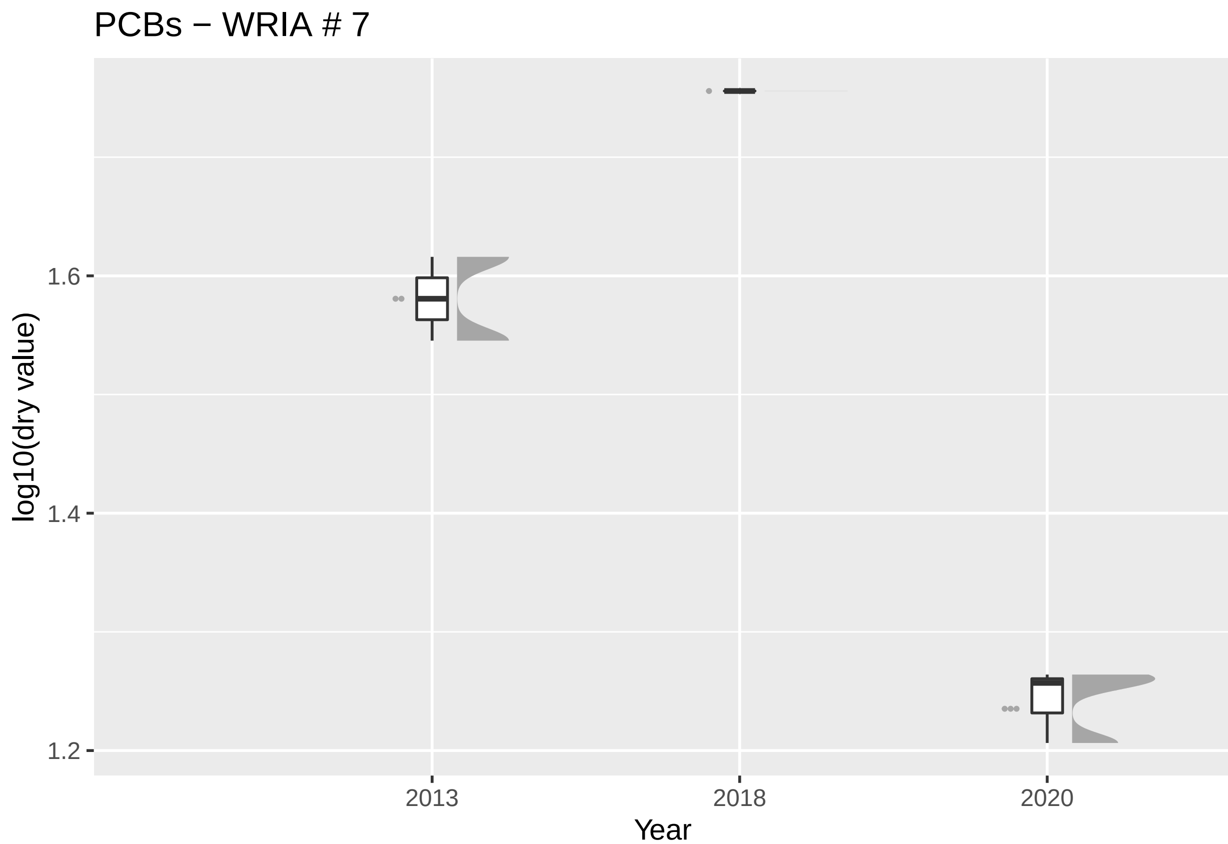
```
##
## $‘17‘
```


PCBs − WRIA # 17
log10(dry value)
1.6
1.4
1.2
1.0
2013
2016
2018
2020
Year

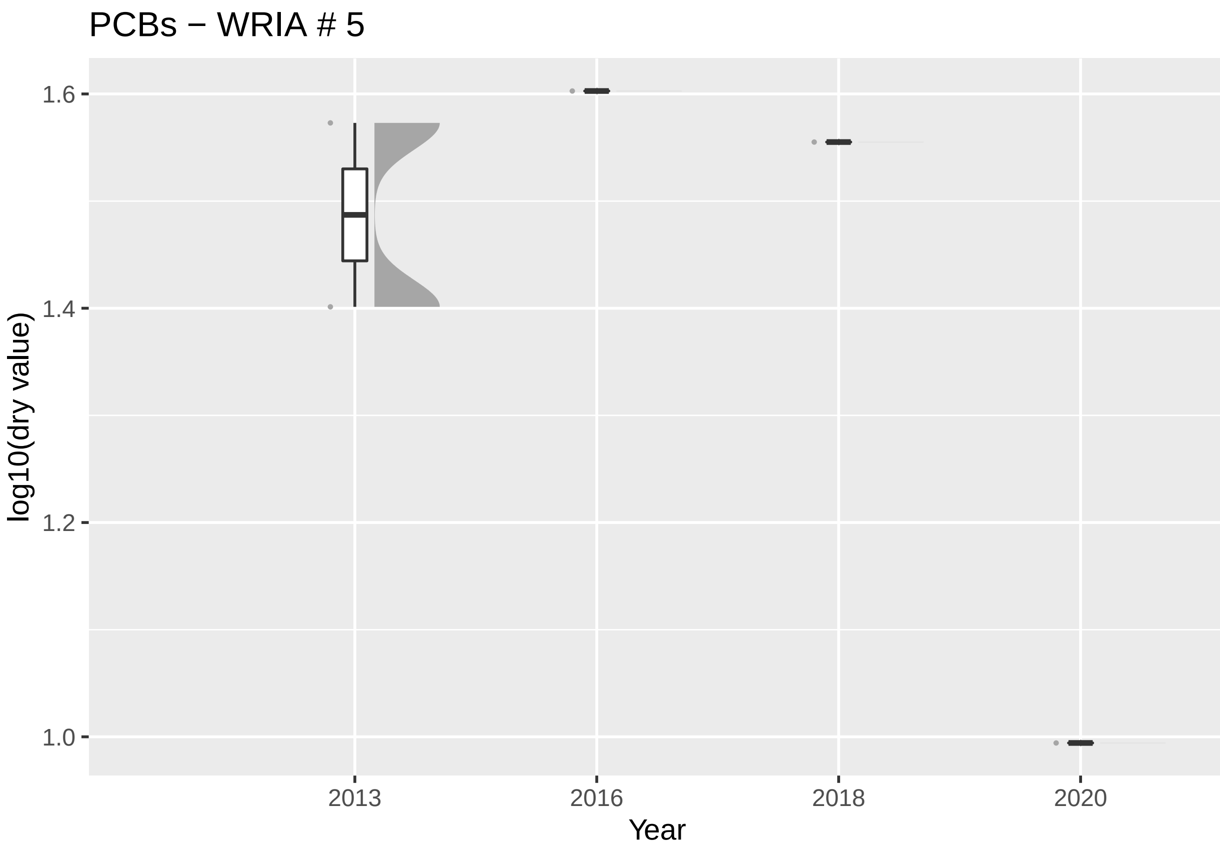
```
##
## $‘6‘
```



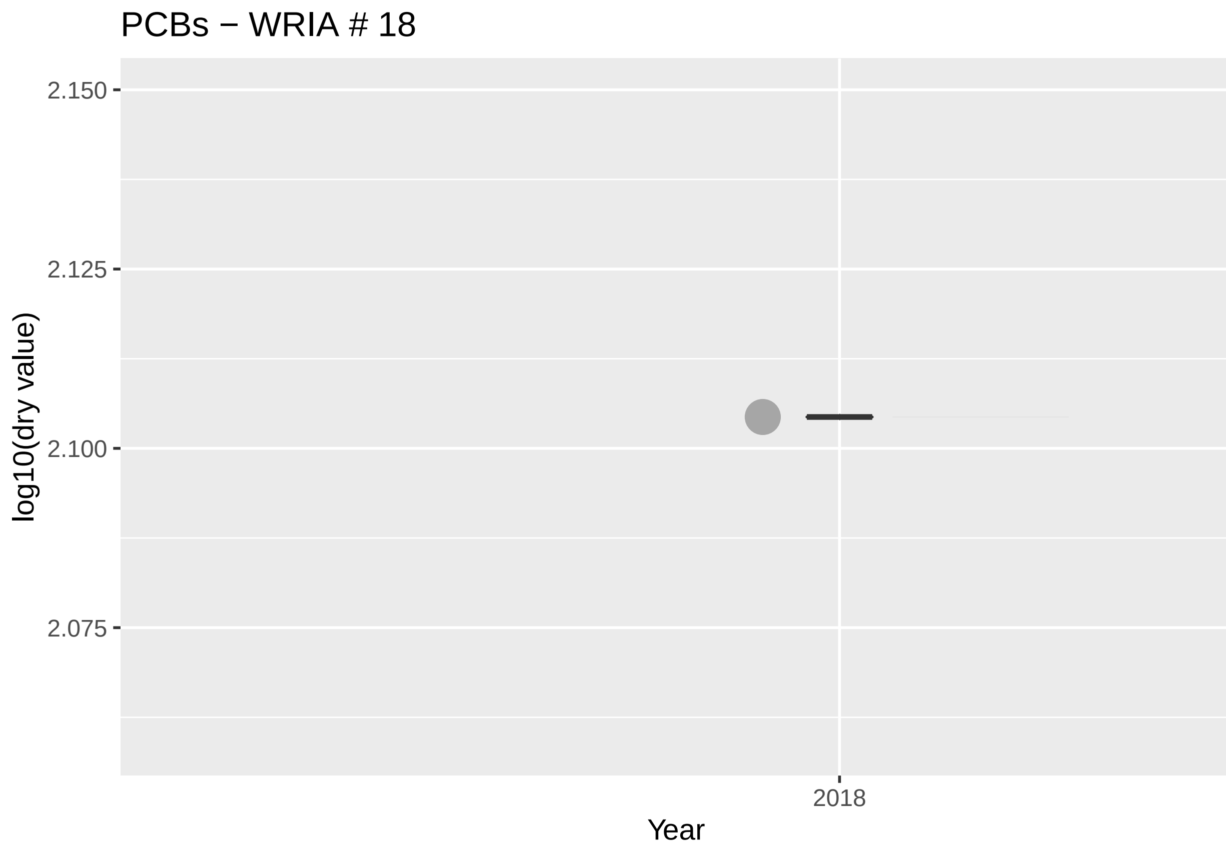

```
##
## $‘7‘
```



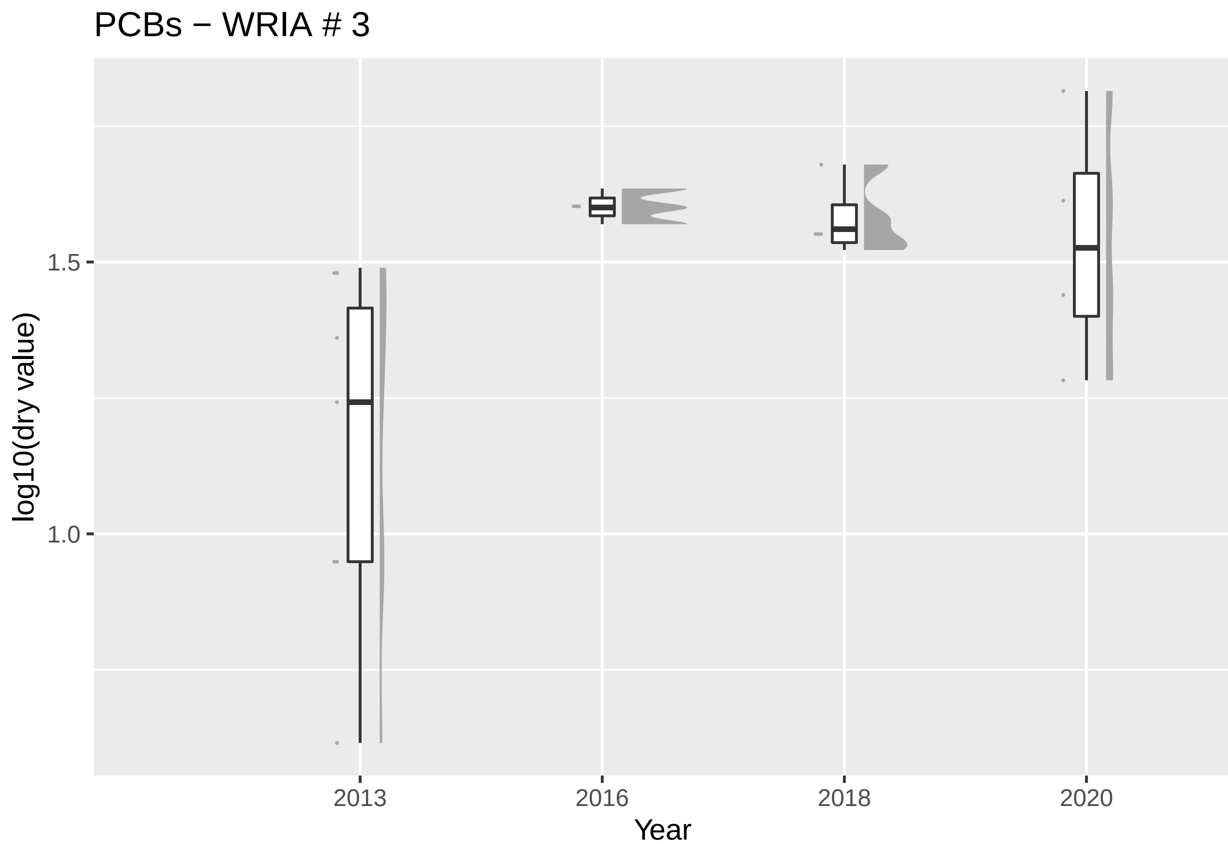

```
##
## $`5`
```


PCBs − WRIA # 5
log10(dry value)
1.6
1.4
1.2
1.0
2013
2016
2018
2020
Year

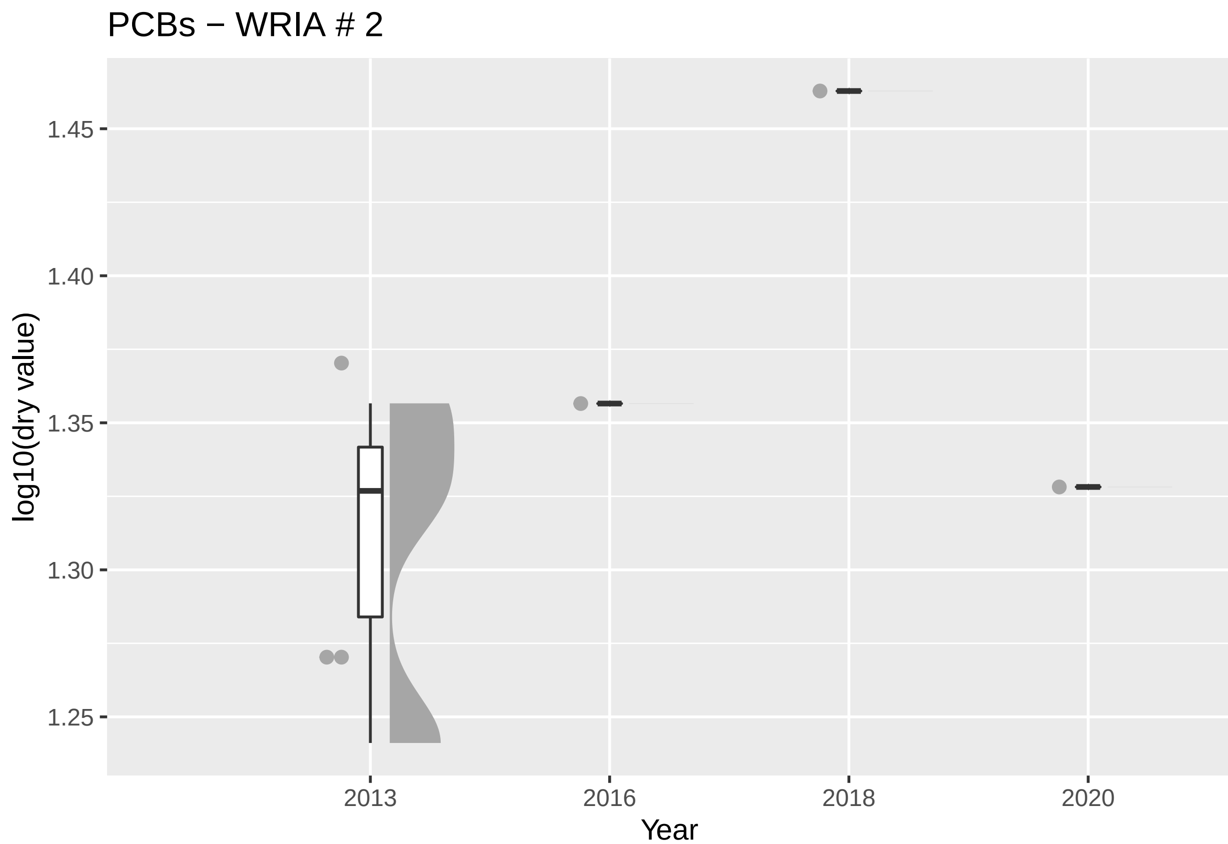
```
##
## $`18`
```



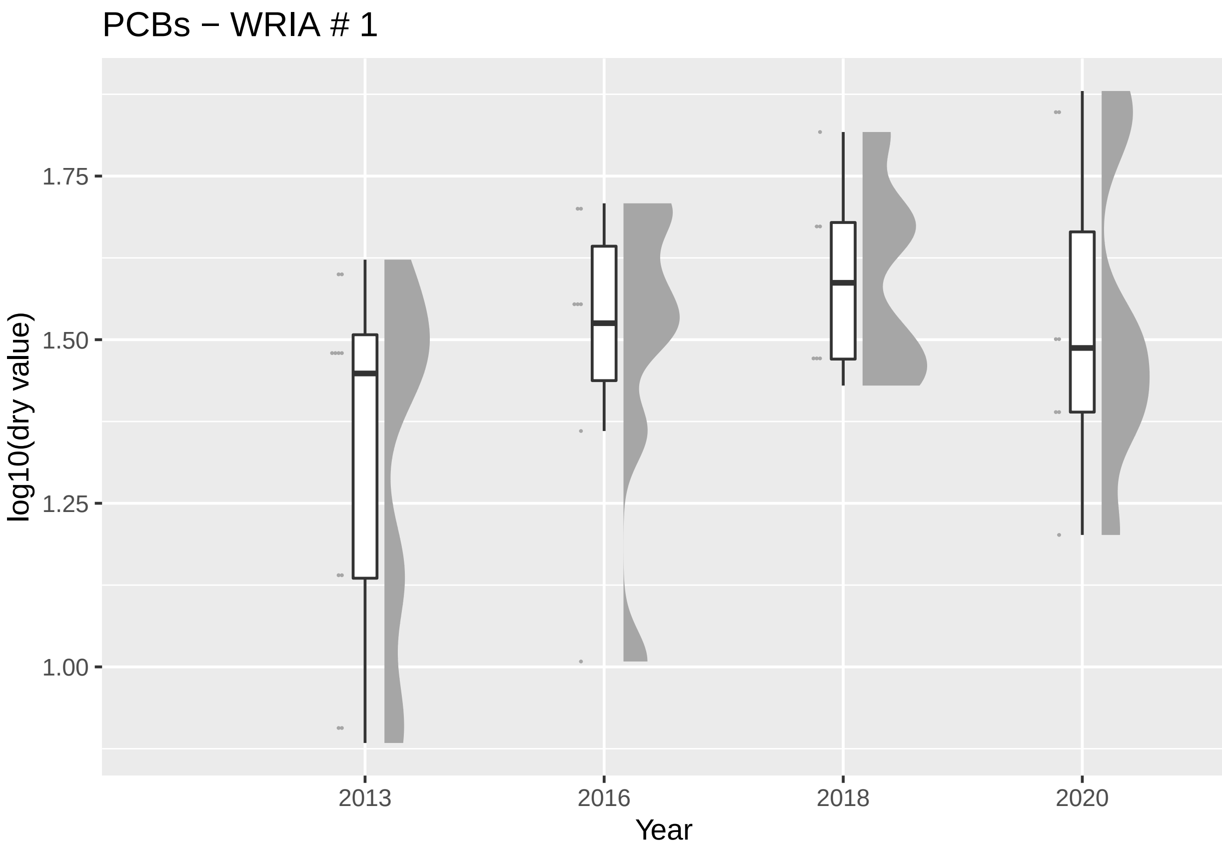

```
##
## $‘3‘
```




```
##
## $`2`
```


PCBs − WRIA # 2
1.45
1.40
1.35
1.30
1.25
log10(dry value)
2013
2016
2018
2020
Year

```
##
## $`1`
```


PCBs − WRIA # 1
log10(dry value)
1.75
1.50
1.25
1.00
2013
2016
2018
2020
Year