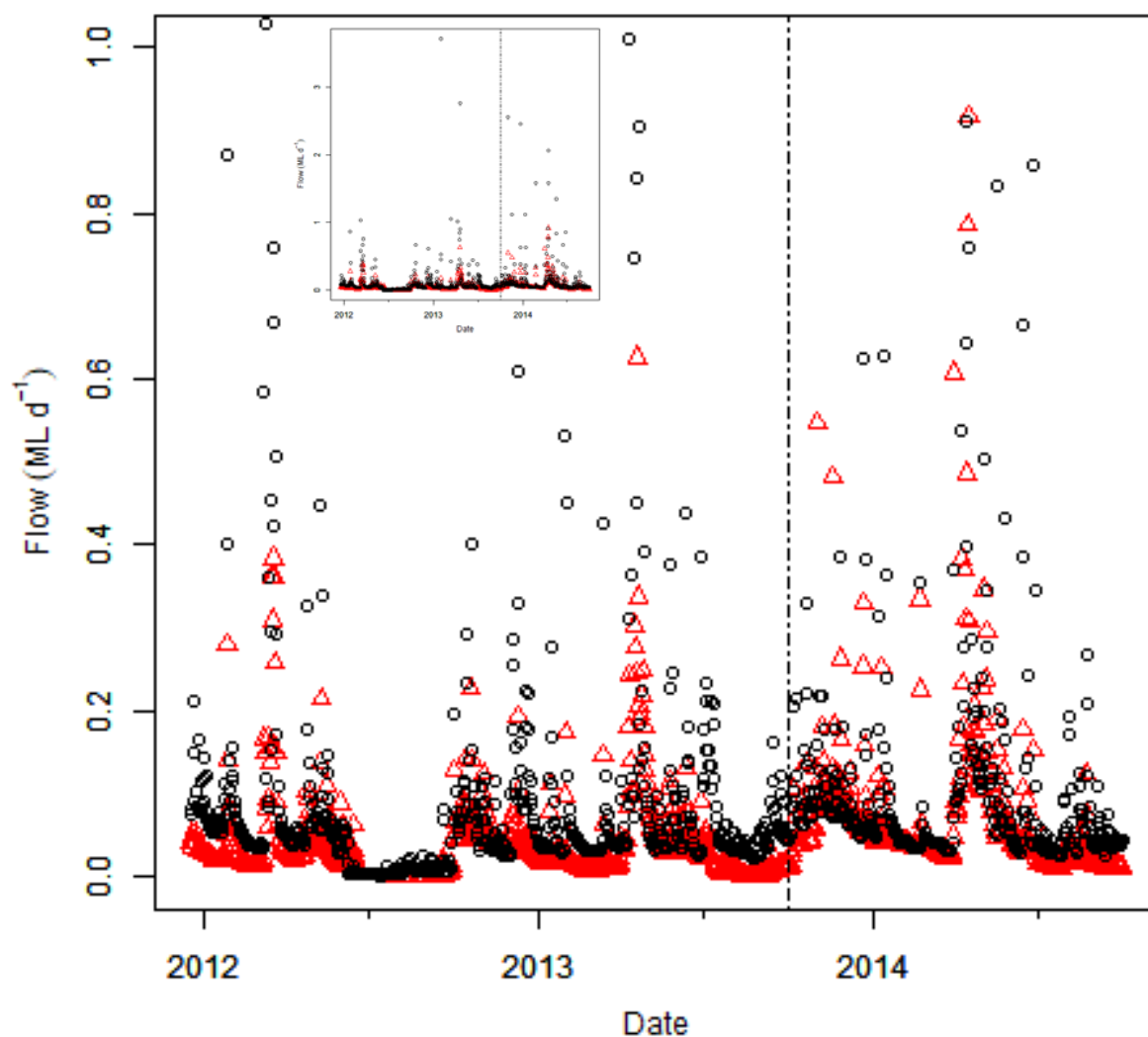
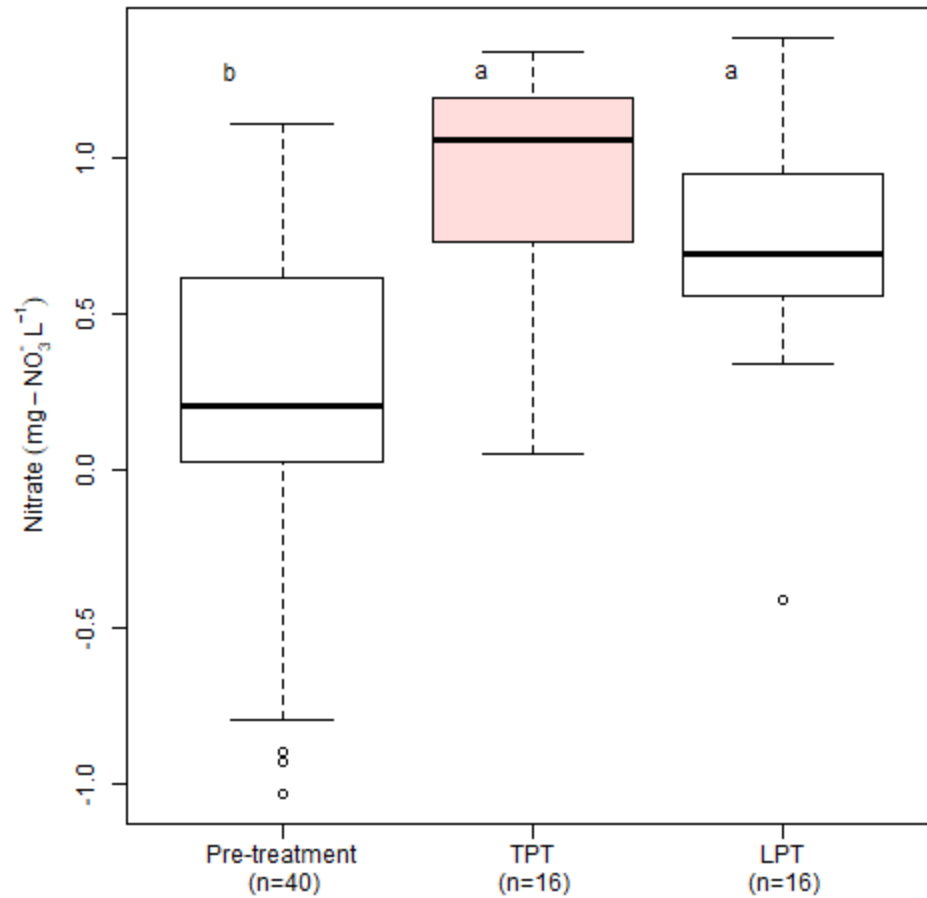


- 1 SUPPORTING INFORMATION
- 2 This material is available free of charge via the Internet.



- 3
- 4 Figure S1: Flow from the treatment and reference tributaries. The small inset shows all the daily
- 5 average flows, while the larger image is showing average daily flows less than 1 ML d⁻¹. There
- 6 was not a statistically significant difference in flows between the tributaries over the period of
- 7 the study.



8

9 Figure S2: Difference in NO_3^- concentrations between limed and reference sites (limed –
 10 reference). TPT and LPT periods were significantly higher than during the pre-treatment period,
 11 indicating an increase in NO_3^- at the limed site following addition. Each box indicates the
 12 interquartile data range, the whiskers indicate the 95% central data range, and outliers are
 13 indicated by unfilled circle symbols.

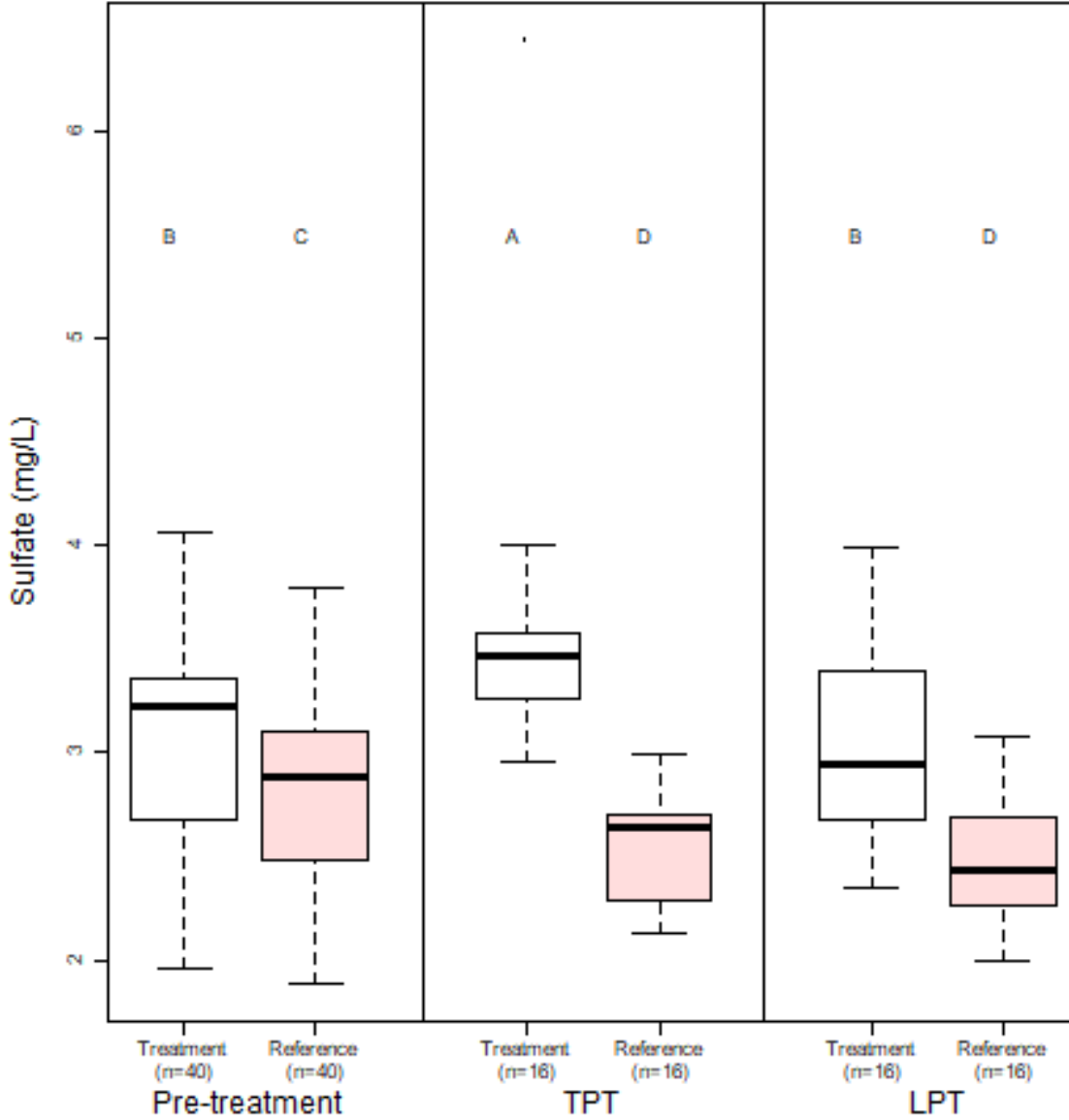


Figure S3: Sulfate (SO_4^{2-} , $\text{mg-SO}_4^{2-} \text{ L}^{-1}$) concentrations from pre-treatment, transitional post treatment (TPT) and longer-term post treatment (LPT) periods for both the limed and reference tributaries. Letter labels indicate significant difference ($\alpha=0.05$). Boxes and whiskers are as described in Fig. S2 caption.

Table S1: Correlations of dissolved organic carbon (DOC), sulfate (SO₄⁻²), ultraviolet absorbance at 254nm (UV254) and specific ultraviolet absorbance (SUVA) with total mercury (THg) and methylmercury (MeHg) at both study sites over the entire period of study. Only THg had statistically significant correlations with DOC, UV254 and SUVA. There were no significant correlations with MeHg.

	THg				MeHg			
	Reference		Treatment		Reference		Treatment	
	p-value	adj-R2	p-value	adj-R2	p-value	adj-R2	p-value	adj-R2
DOC	<0.001	0.627	<0.001	0.900	0.537	-0.44	0.813	-0.067
Sulfate	0.332	0.001	0.566	-0.040	0.231	0.040	0.189	0.056
Nitrate	0.038	0.238	0.515	-0.034	0.305	0.010	0.541	-0.042
UV254	0.001	0.528	<0.001	0.900	0.865	-0.074	0.848	-0.069
SUVA	0.321	0.004	0.002	0.412	0.136	0.098	0.662	-0.056

26 **Table S2:** Each analyte was tested with a standard ANOVA during the pre-treatment period to determine
 27 if significant differences existed between the treatment and reference tributaries. All available data was
 28 used for this test, in addition to paired samples. No significant difference was found in pH, however the
 29 other analytes were higher in the limed tributary. The power for all Hg related tests during this period is
 30 low, so no differences were detected, despite significantly higher DOC at the limed site.

Analyte	p-value
pH	0.111
DOC (mg L ⁻¹)	0.013
UV254 (cm ⁻¹)	0.002
SUVA (mg-C ⁻¹ m ⁻¹)	<0.001
Sulfate (mg-SO ₄ ²⁻ L ⁻¹)	0.034
Nitrate (mg-NO ₃ ⁻ L ⁻¹)	0.542
THg (ng L ⁻¹)	0.48
MeHg (ng L ⁻¹)	0.394
MeHg/THg (%)	0.8
THg:DOC (µg g ⁻¹)	0.84
DOC flux	<0.001
SO ₄ ²⁻ Flux	<0.001

31

32 Table S3: Results of LMS fixed model ANOVA tests. The linear model slope was significantly
 33 different from zero for all but %MeHg and MeHg:DOC. Where interaction was significant
 34 (Source*Period $p < 0.2$, marginal $p < 0.3$) BACI tests were used to determine differences between
 35 time periods. When this interaction was not present, the main effect of Source and Period were
 36 evaluated ($\alpha = 0.05$).

Parameter	Linear Model	Source	Period	Source*Period
pH	<0.001	<0.001	<0.001	<0.001
DOC	<0.001	<0.001	<0.001	<0.001
UV ₂₅₄	<0.001	<0.001	<0.001	<0.001
SUVA	<0.001	<0.001	<0.001	<0.001
SO ₄ ²⁻	<0.001	<0.001	0.014	<0.001
NO ₃ ⁻	0.001	0.006	0.013	0.165
THg	<0.001	<0.001	0.013	0.215
MeHg	0.06	0.009	0.174	0.757
%MeHg	0.251	0.53	0.063	0.792
THg:DOC	0.002	0.004	0.01	0.132
MeHg:DOC	0.257	0.207	0.125	0.670

37

Table S4: Before-after control-impact (BACI) test results comparing the difference in paired treatment measurements (limed – reference) for each analyte of interest among the three sampling periods. Few differences were detected for Hg metrics due to low sample size, despite a significant correlation between THg and DOC. Estimate values indicate which time period had the larger difference between sites. A positive estimate indicates the earlier time period was larger (eg. TPT>LPT).

	pH		DOC		UV254		SUVA		Sulfate		Nitrate	
	Estimate	p-value	Estimate	p-value	Estimate	p-value	Estimate	p-value	Estimate	p-value	Estimate	p-value
pre v LPT	-0.93	<0.001	-1.90	<0.001	-0.10	<0.001	-0.35	<0.001	-0.57	<0.001	-0.51	0.001
pre v TPT	-2.15	<0.001	-4.07	<0.001	-0.19	<0.001	-0.30	<0.001	-0.91	<0.001	-0.74	<0.001
TPT v LPT	1.22	<0.001	2.17	<0.001	0.10	0.003	-0.05	0.494	0.33	0.030	0.23	0.187
	MeHg		THg		MeHg/THg		THg:DOC		MeHg:DOC			
	Estimate	p-value	Estimate	p-value	Estimate	p-value	Estimate	p-value	Estimate	p-value		
pre v LPT	-0.03	0.431	-0.62	0.363	-0.01	0.661	-0.11	0.015	-0.01	0.668		
pre v TPT	-0.02	0.533	-1.47	0.043	0.01	0.820	-0.06	0.179	<0.01	0.913		
TPT v LPT	-0.01	0.819	0.85	0.134	-0.02	0.370	-0.06	0.108	-0.01	0.343		