

Supplemental Tables

Table S1 Owners of oil sands facilities, start-up years, and production capacity; obtained from Oil Sands Magazine (<https://www.oilsandsmagazine.com/projects/bitumen-production>); accessed November 2020.

| Owner | Facility | Type | Start-up year | Production capacity (BPD) | Notes |
|-------------------------------------|--------------------------------------|---------|---------------|---------------------------|------------------|
| Athabasca | Hangingstone | In situ | 2015 | 12,000 | |
| | Leismer | In situ | 2010 | 20,000 | |
| International Petroleum Corporation | Blackrod | In situ | 2011 | 500 | |
| Canadian Natural | Horizon | Mine | 2009 | 294,000 | |
| | Muskeg River | Mine | 2002 | 175,000 | |
| | Jackpine | Mine | 2010 | 145,000 | |
| | Kirby | In situ | 2013 | 80,000 | |
| | Wolflake/ Primrose/ Burnt Lake | In situ | 1985 | 120,000 | |
| | Peace River/ Carmon Creek | In situ | 1986 | 12,500 | |
| Cenovus | Christina Lake | In situ | 2002 | 260,000 | |
| | Foster Creek | In situ | 2001 | 180,000 | |
| CNOOC | Nexen | In situ | 2008 | 92,000 | |
| CONA | Lindbergh | In situ | 2012/2015 | | pilot/commercial |
| Connacher | Great Divide | In situ | 2008 | 20,000 | |
| Conocophillips | Surmont | In situ | 2007 | 148,000 | |
| Greenfire | Hangingstone Demo | In situ | 1999 | 10,000 | |
| Harvest | Blackgold | In situ | 2018 | 10,000 | |
| Husky | Sunrise | In situ | 2015 | 60,000 | |
| | Tucker Lake | In situ | 2006 | 30,000 | |
| Imperial | Kearl | Mine | 2013 | 220,000 | |
| | Cold Lake | In situ | 1975/1985 | | pilot/commercial |
| JACOS | Hangingstone EXP | In situ | 2017 | 20,000 | |
| MEG | Christina Lake | In situ | 2008 | 100,000 | |
| OSUM | Orion | In situ | 2007 | 18,000 | |
| Petrochina | MacKay River | In situ | 2017 | 35,000 | |
| Suncor | Basemine | Mine | 1967 | N/A | |
| | Millenium | Mine | 2001 | 180,000 | |
| | Steepbank | Mine | 2010 | 150,000 | |
| | Fort Hills | Mine | 2018 | 194,000 | |
| | MacKay River | In situ | 2002 | 38,000 | pilot in 1983 |
| | Firebag | In situ | 2004 | 215,000 | |
| Sunhsine | West Ells | In situ | 2015 | 5,000 | |
| Syncrude | Mildred Lake | Mine | 1978 | 150,000 | |
| | Aurora North | Mine | 2001 | 225,000 | |

Table S2 History of activities in the Muskeg basin estimated from satellite imagery.

| Year | Event |
|-----------|----------------------------------------------------------------------------------------------------------|
| 1974-1975 | Shell test pit constructed on Lease 13 |
| 1978-1979 | Hwy 63. Bridge construction across Athabasca River |
| | Extension of Hwy. 63 and Canterra Road. |
| | Air strip on/near eventually Aurora North Lease |
| | Land clearance near air strip |
| 1981 | Seismic lines apparent; original date not clear |
| | Extension of Canterra Road |
| | land clearance |
| | Logging camps? |
| 1982-1996 | Logging (e.g., south of Kearn Lake) |
| | Road Construction |
| | Land clearance |
| 1997 | Construction of Aurora North begins |
| 1998 | Construction of Muskeg River Mine begins |
| 2000 | Expansion of MRM into Sharkbite lease (wedge west of Jackpine Creek and south of Muskeg River) |
| 2001 | Suncor Firebag construction begins |
| | Aurora North begins commercial operations |
| 2002 | Utility corridor to Suncor Firebag |
| | MRM begins commercial operations |
| 2003 | MRM External tailings facility filled |
| 2004 | Suncor Firebag begins commercial operations |
| 2006 | Construction of Jackpine Mine begins |
| | Hammerstone quarry construction begins |
| 2007 | Husky Sunrise construction begins |
| | Firebag aerodrome construction |
| | construction of aerodrome on Sharkbite lease (MRM) |
| 2008 | Kearn begins construction |
| | land clearance for Steepbank mine crosses into Muskeg drainage |
| 2009 | Excavation of JPM compensation lake |
| | Excavation of Kearn compensation lake |
| | Road construction south from Hammerstone quarry |
| | Minor land clearing on Aurora South Lease |
| 2010 | JPM External tailings facility filled |
| | JPM begins commercial operation |
| | North Steepbank Mine begins commercial operation |
| | Intensification of MRM activity in Sharkbite Lease |
| 2011 | Road construction and minor land clearing for Jackpine Mine Expansion Lease |
| 2012 | Land clearing for Imperial Aspen |
| 2013 | Fort Hills construction crosses into Muskeg drainage |
| | Imperial Kearn begins commercial operation |
| 2015 | Husky Sunrise begins commercial operations |
| 2013-2019 | Progression of mine operations (e.g., expansion of mine pits) and additional activities throughout basin |

Table S3 History of activities in the Steepbank basin estimated from satellite imagery.

| Year | Event |
|-----------|--------------------------------------------------------------------------------|
| 1996 | Forestry |
| 1997 | Land clearing for Millenium mine, but minor within Steepbank drainage |
| 1998-1999 | Land clearance and more expansion into Steepbank drainage |
| 2001 | Road construction |
| | Forestry |
| | Road to Suncor Firebag from Muskeg drainage (North Steepbank River) |
| | Millenium mine begins commerical operations |
| 2002 | Utility corridor to Suncor Firebag |
| 2003 | Minor expansion of MM into Steepbank drainage |
| 2004 | Land disturbance from Millenium to Steepbank River |
| | Suncor Firebag begins commerical operation |
| 2005 | Expansion of 2004 land disturbance of MM |
| 2006 | Expansion of MM |
| 2007-2008 | Land clearing for North Steepbank Mine |
| 2010 | North Steepbank Mine begins commercial operations |
| 2015 | Husky Sunrise begins commercial operations |
| 2013-2019 | Land clearance for MM |
| | Expansion of NSM since 2008 |
| | Continued mine development, including pit expansions within existing footprint |

Table S4 History of activities in the McLean Creek basin estimated from satellite imagery.

| Year | Event |
|-----------|-----------------------------------------------------|
| 1996 | Land Clearance |
| 1998 | forestry/land clearance |
| 2000 | land clearance for Millenium Mine |
| 2001 | Construction; tailings pond (8A and 8B) |
| | Commercial operations of Millenium Mine begin |
| 2003 | Filling 8A and 8B |
| 2004 | Land clearing for South Tailings Pond (STP) |
| 2005 | begin construction of STP |
| | Diversion of McLean Creek |
| 2007 | Fill STP |
| 2011 | Millenium mine expansion into Mclean Creek drainage |
| 2012-2019 | Continued development of Millenium mine |

Table S5 History of activities in the MacKay River basin estimated from satellite imagery.

| Year | Event |
|-----------|--------------------------------------------------------------------------------------------|
| <1972 | Bridge to Fort MacKay |
| 1975 | Forestry |
| 1984 | Land clearing at Suncor Dover; future Suncor MacKay River in situ (MRIS) site |
| | Forestry |
| 1989 | Construction of Southwest Sand Storage facility at Mildred Lake (in Beaver River drainage) |
| | Forestry |
| 1991 | Mildred Lake Settling Basin expansion; NW |
| 1993 | SWSS expansion crosses into MacKay drainage |
| 1994-1995 | Forestry |
| 2001 | Expansion of MRIS |
| 2002 | SWSS expansion |
| | Construction of road northwards from MRIS into Ells drainage |
| | Commercial operation of MRIS begins |
| 2003 | Construction of CNRL Horizon Hwy in MacKay drainage |
| 2004 | Forestry |
| | MRIS expansion |
| | Horizon construction begins |
| 2008 | Forestry |
| 2009 | Mildred Lake expansion into MacKay drainage |
| 2011 | Road construction to PetroChina MacKay River lease |
| | Expansion of ML |
| 2012 | Expansion of ML |
| | Road construction to West Ells lease |
| | In situ construction (West Ells and PetroChina Mackay River) |
| 2013-2014 | Expansion of in situ facilities |
| 2015 | Commercial operation of Sunshine West Ells begins |
| 2017 | Commercial operation of Petrochina MacKay River begins |
| 2017-2019 | Expansion of existing facilities |

Table S6 History of activities in the Ells River basin estimated from satellite imagery.

| Year | Event |
|-----------|---------------------------------------------------------|
| <1984 | Seismic lines present in lower watershed |
| 1987 | logging Road constructed in south of basin |
| 1992 | Land Clearance straddling divide with Tar River |
| 1998 | Land clearance |
| 2002 | Road from MacKay River in situ constructed |
| 2003 | Horizon Hwy constructed |
| | construction of camps |
| | drilling activity |
| 2005 | Exploration activities |
| 2007 | Drilling |
| 2008 | Site Preparation for JNM begins |
| 2009 | Horizon begins commercial operations in Tar basin |
| 2010-2011 | Land clearance |
| 2012 | Construction of Joslyn North |
| | Land clearance NW of Horizon ETF |
| 2014 | Land clearance for JN; project abandoned |
| 2020 | Site prep for Horizon South (former Joslyn North lease) |

Table S7 History of activities in the Tar River basin estimated from satellite imagery.

| Year | Event |
|-----------|-----------------------------------------------------|
| <1984 | Seismic lines |
| 1991 | Logging |
| 1992 | Land clearing at Ells divide |
| 2002 | Road construction |
| 2003 | Horizon Hwy constructed |
| 2004 | major land clearance |
| 2005 | Horizon construction begins |
| 2007 | Construction of Horizon Lake and interception ditch |
| 2008 | Filling of Horizon ETF |
| | Begin diversion of Tar River |
| 2009 | Commercial operations of Horizon begin |
| | Complete diversion of Tar River |
| 2009-2019 | Expansion of Horizon |

Table S8 Summary Tables for tributary site GEEs.

| Site | Variable | Estimate (ln) | Std.err | Wald | Pr(> W) |
|------|---------------|---------------|----------|----------|----------|
| ELR1 | (Intercept) | 4.597862 | 1.26E-05 | 1.32E+11 | <0.001 |
| | Alkalinity | 0.008637 | 1.70E-08 | 2.58E+11 | <0.001 |
| | condtypeD1 | 0.020676 | 1.41E-05 | 2149098 | <0.001 |
| | condtypeD2 | -0.47652 | 1.83E-05 | 6.78E+08 | <0.001 |
| | Discharge | -0.00319 | 6.26E-08 | 2.6E+09 | <0.001 |
| | Precipitation | 0.00178 | 1.14E-08 | 2.46E+10 | <0.001 |
| JP1 | (Intercept) | 4.7319 | 2.25E-06 | 4.40E+12 | <0.001 |
| | Alkalinity | 0.005851 | 5.50E-09 | 1.13E+12 | <0.001 |
| | condtypeD1 | 0.016348 | 1.66E-06 | 97106398 | <0.001 |
| | condtypeD2 | 0.083659 | 3.24E-06 | 6.68E+08 | <0.001 |
| | Discharge | -0.00412 | 2.07E-07 | 3.95E+08 | <0.001 |
| | Precipitation | -8.70E-05 | 9.34E-09 | 86810567 | <0.001 |
| MCC1 | (Intercept) | 4.67294 | 4.34E-05 | 1.16E+10 | <0.001 |
| | Alkalinity | 0.007862 | 1.41E-07 | 3.13E+09 | <0.001 |
| | Discharge | 0.015314 | 1.60E-06 | 92004324 | <0.001 |
| | Precipitation | -0.00729 | 5.48E-07 | 1.77E+08 | <0.001 |
| MUR1 | (Intercept) | 5.309609 | 3.40E-05 | 2.43E+10 | <0.001 |
| | Alkalinity | 0.003522 | 1.27E-07 | 7.73E+08 | <0.001 |
| | Discharge | -0.01515 | 1.30E-06 | 1.35E+08 | <0.001 |
| | Precipitation | 0.002878 | 2.52E-08 | 1.31E+10 | <0.001 |
| | condtypeD1 | -0.02702 | 2.40E-06 | 1.26E+08 | <0.001 |
| | condtypeD2 | -0.04291 | 3.20E-06 | 1.8E+08 | <0.001 |
| MUR2 | (Intercept) | 4.92481 | 5.63E-06 | 7.64E+11 | <0.001 |
| | Alkalinity | 0.005436 | 2.89E-09 | 3.54E+12 | <0.001 |
| | condtypeD1 | -0.08674 | 1.72E-06 | 2.55E+09 | <0.001 |
| | condtypeD2 | -0.14504 | 1.20E-06 | 1.45E+10 | <0.001 |
| | Discharge | 0.00301 | 1.76E-08 | 2.91E+10 | <0.001 |
| | Precipitation | -0.00064 | 9.80E-09 | 4.28E+09 | <0.001 |
| MUR3 | (Intercept) | 4.9627 | 3.12E-07 | 2.53E+14 | <0.001 |
| | Alkalinity | 0.004742 | 2.95E-09 | 2.58E+12 | <0.001 |
| | condtypeD1 | -0.01807 | 2.49E-07 | 5.28E+09 | <0.001 |
| | condtypeD2 | -0.02597 | 2.68E-07 | 9.36E+09 | <0.001 |
| | Discharge | 0.000852 | 7.11E-09 | 1.44E+10 | <0.001 |
| | Precipitation | -0.00036 | 1.32E-08 | 7.63E+08 | <0.001 |
| MUR7 | (Intercept) | 5.030801 | 1.23E-05 | 1.66E+11 | <0.001 |
| | Alkalinity | 0.004128 | 5.11E-08 | 6.53E+09 | <0.001 |
| | condtypeD1 | -0.02948 | 1.28E-06 | 5.34E+08 | <0.001 |
| | condtypeD2 | -0.15637 | 4.48E-06 | 1.22E+09 | <0.001 |
| | Discharge | -0.00079 | 2.86E-07 | 7569624 | <0.001 |
| | Precipitation | 3.62E-06 | 1.75E-08 | 43099.77 | <0.001 |
| STR1 | (Intercept) | 4.799237 | 1.63E-05 | 8.67E+10 | <0.001 |
| | Alkalinity | 0.005385 | 7.06E-08 | 5.81E+09 | <0.001 |
| | condtypeD1 | 0.026022 | 2.22E-06 | 1.37E+08 | <0.001 |
| | condtypeD2 | -0.03887 | 6.12E-07 | 4.03E+09 | <0.001 |
| | Discharge | -0.00593 | 6.40E-07 | 85795494 | <0.001 |
| | Precipitation | 0.002354 | 1.31E-07 | 3.23E+08 | <0.001 |
| TAR1 | (Intercept) | 5.289424 | 2.36E-06 | 5.03E+12 | <0.001 |
| | Alkalinity | 0.004813 | 1.96E-08 | 6.01E+10 | <0.001 |
| | condtypeD1 | 0.084358 | 2.83E-07 | 8.86E+10 | <0.001 |
| | Discharge | -0.01298 | 4.59E-07 | 7.98E+08 | <0.001 |
| | Precipitation | 0.001866 | 2.10E-07 | 79143429 | <0.001 |

Table S9 Summary Tables for large river site GEEs.

| Site | Variable | Estimate (ln) | Std.err | Wald | Pr(> W) |
|---------|---------------|---------------|----------|----------|----------|
| AR10/11 | (Intercept) | 4.878364 | 1.27E-05 | 1.47E+11 | <0.001 |
| | Alkalinity | 0.008246 | 8.11E-08 | 1.03E+10 | <0.001 |
| | condtypeD1 | 0.006761 | 2.79E-06 | 5858215 | <0.001 |
| | Discharge | -0.00018 | 1.12E-08 | 2.6E+08 | <0.001 |
| | Precipitation | 0.000543 | 8.33E-08 | 42448434 | <0.001 |
| AR3 | (Intercept) | 4.982466 | 2.19E-05 | 5.18E+10 | <0.001 |
| | Alkalinity | 0.006269 | 1.37E-07 | 2.1E+09 | <0.001 |
| | condtypeD1 | -0.16573 | 4.76E-06 | 1.21E+09 | <0.001 |
| | condtypeD2 | -0.0636 | 2.38E-06 | 7.15E+08 | <0.001 |
| | Discharge | -0.00013 | 1.33E-08 | 97658705 | <0.001 |
| | methodD1 | 0.003715 | 4.19E-06 | 784840.9 | <0.001 |
| | methodD2 | 0.008235 | 1.66E-06 | 24514755 | <0.001 |
| | Precipitation | 0.001575 | 4.28E-08 | 1.36E+09 | <0.001 |
| Clear1 | (Intercept) | 5.208979 | 6.23E-06 | 6.98E+11 | <0.001 |
| | Alkalinity | 0.005542 | 3.76E-08 | 2.18E+10 | <0.001 |
| | condtypeD1 | 0.204583 | 2.10E-05 | 94969037 | <0.001 |
| | condtypeD2 | 0.10348 | 5.69E-07 | 3.31E+10 | <0.001 |
| | Discharge | -0.0337 | 2.98E-10 | 1.28E+16 | <0.001 |
| | Precipitation | 0.002544 | 9.47E-08 | 7.22E+08 | <0.001 |
| PR3 | (Intercept) | 4.802659 | 1.70E-05 | 7.95E+10 | <0.001 |
| | Alkalinity | 0.00551 | 1.53E-07 | 1.3E+09 | <0.001 |
| | Discharge | 0.002191 | 1.05E-07 | 4.31E+08 | <0.001 |
| | Precipitation | 0.000348 | 5.16E-08 | 45507309 | <0.001 |
| PR4 | (Intercept) | 4.702327 | 5.63E-06 | 6.98E+11 | <0.001 |
| | Alkalinity | 0.007855 | 6.20E-08 | 1.6E+10 | <0.001 |
| | Discharge | 0.001646 | 3.79E-08 | 1.88E+09 | <0.001 |
| | Precipitation | -0.00223 | 6.96E-08 | 1.02E+09 | <0.001 |
| SR1 | (Intercept) | 4.808375 | 2.07E-06 | 5.39E+12 | <0.001 |
| | Alkalinity | 0.006343 | 3.60E-08 | 3.11E+10 | <0.001 |
| | Discharge | 0.000823 | 2.41E-08 | 1.17E+09 | <0.001 |
| | Precipitation | -0.00083 | 2.14E-08 | 1.52E+09 | <0.001 |

Table S10 Correlations of data estimated by the GEE using the 'AR1' working correlation structure.

| Site | Data correlations |
|---------|-------------------|
| MUR1 | 0.551 |
| MUR2 | 0.943 |
| MUR3 | 0.217 |
| JP1 | 0.890 |
| MUR7 | 0.235 |
| STR1 | 0.548 |
| MCC1 | 0.190 |
| TAR1 | 0.610 |
| ELR1 | 0.552 |
| MAC1 | 0.437 |
| AR10/11 | 0.680 |
| AR3 | 0.540 |
| Clear1 | 0.712 |
| SR | 0.366 |
| PR4 | 0.487 |
| PR3 | 0.282 |

Supplemental figures

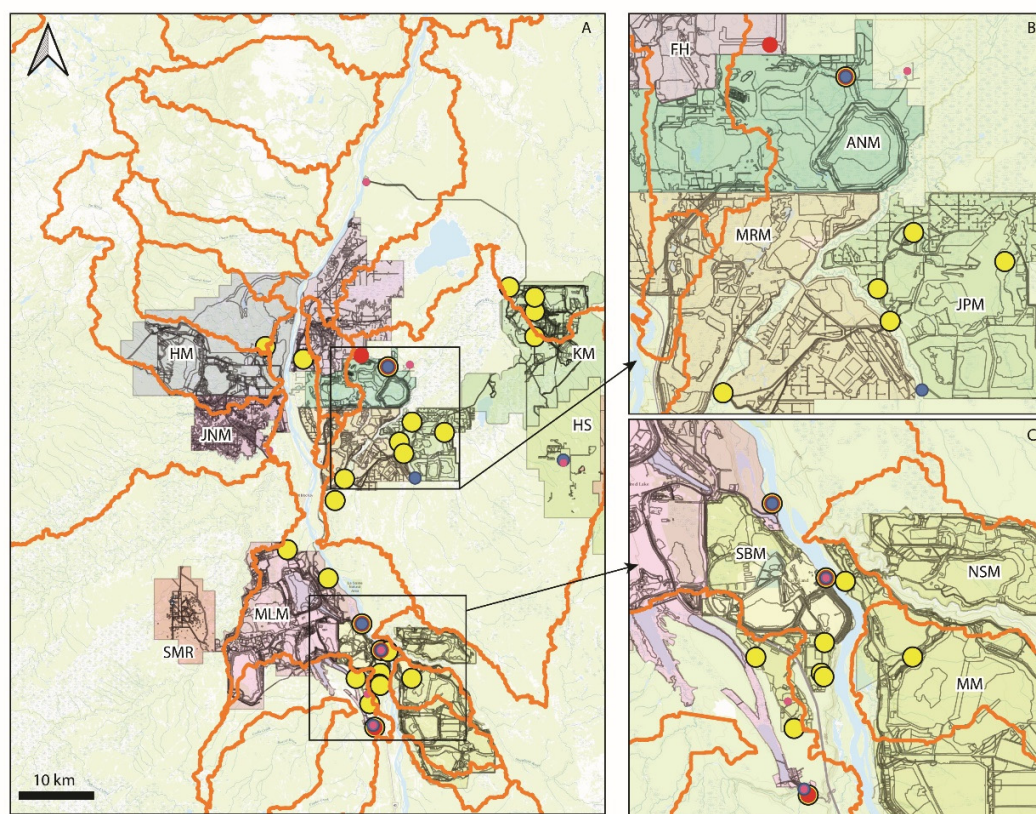


Figure S1 Locations of effluent discharges at oil sands facilities from 2012-2015 (purple, blue, red, yellow, respectively) obtained from Regional Aquatics Monitoring Program (RAMP) reports (RAMP 2016); HM=Horizon Mine; JNM=Joslyn North Mine; SMR= Suncor MacKay River; MLM=Mildred Lake Mine; SBM=Suncor Basemine; MM=Millenium Mine; NSM=North Steepbank Mine; JPM=Jackpine Mine; ANM=Aurora North Mine; MRM=Muskeg River Mine; FH=Fort Hills Mine; HS=Husky Sunrise; KM=Kearl Mine; Oil sands features layer from Oil Sands Information Portal (2016 data layer; <http://osip.alberta.ca/library/Browser>); also shown are the outlines of watershed boundaries (layer obtained from <http://www.ramp-alberta.org/data/map/mapdata.aspx>).

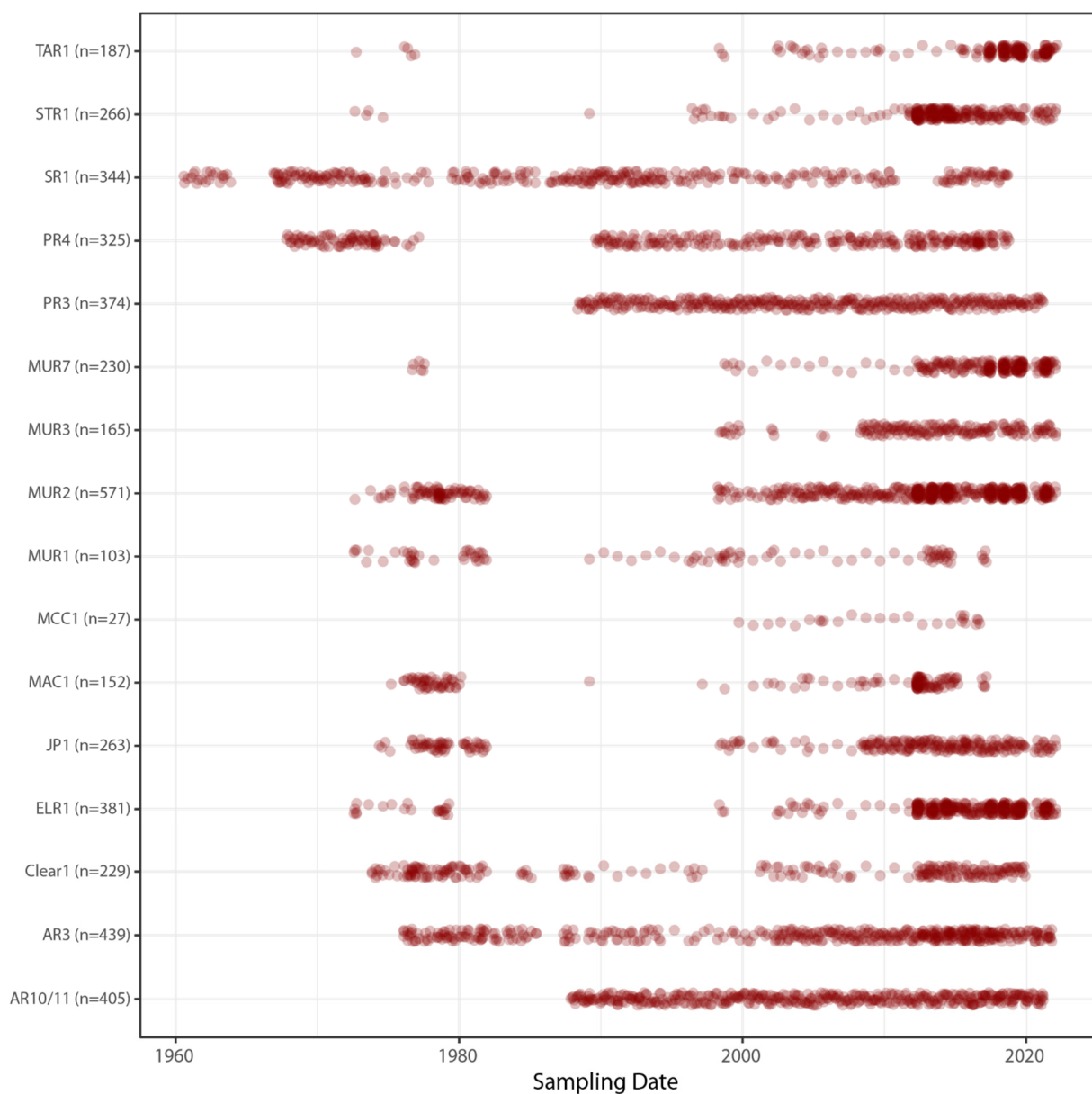


Figure S2 Sample sizes per study location.

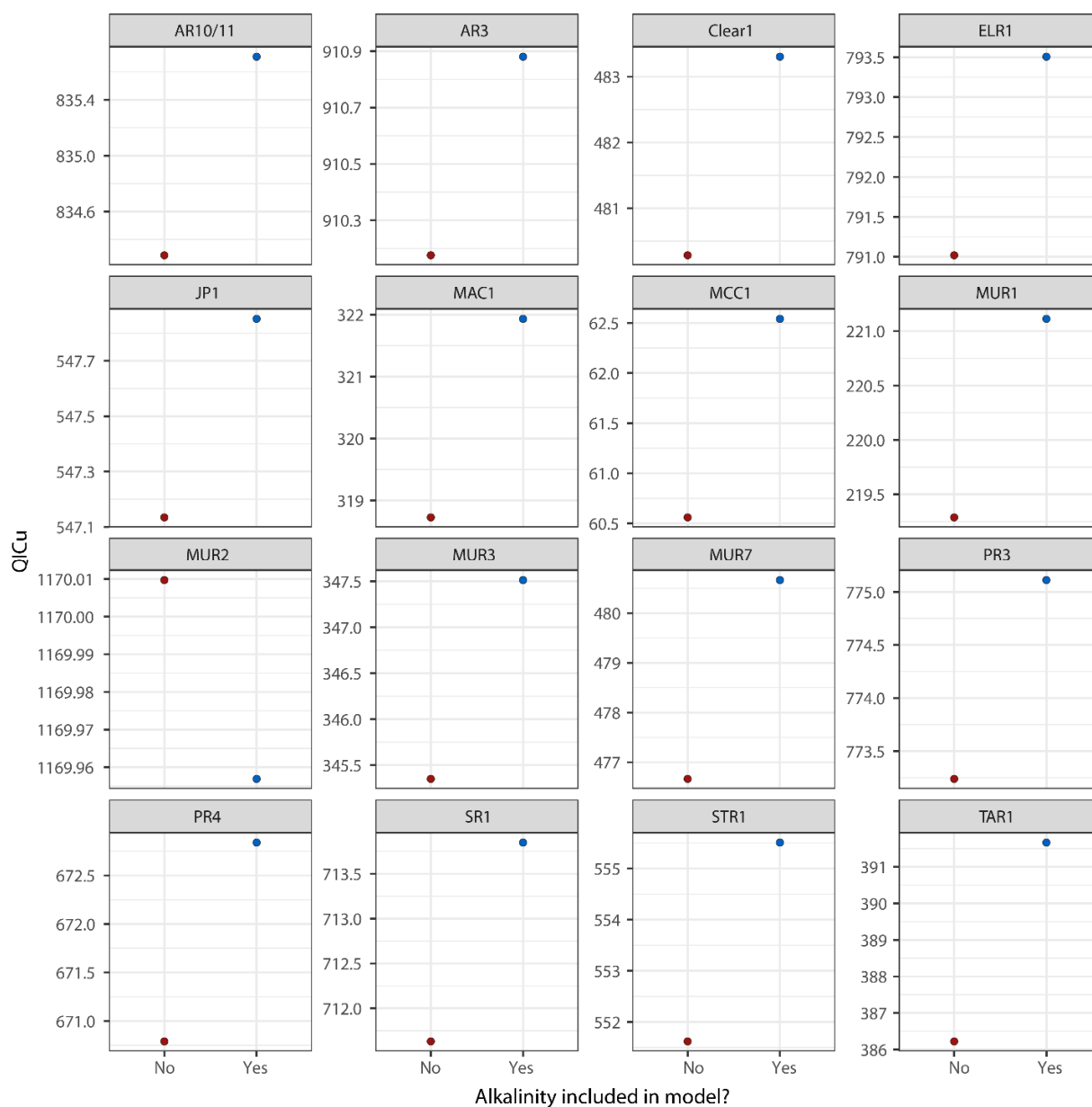


Figure S3 Simplified QIC (QICu) calculated for models with and without alkalinity as an exploratory factor at the 16 sites examined in this study.

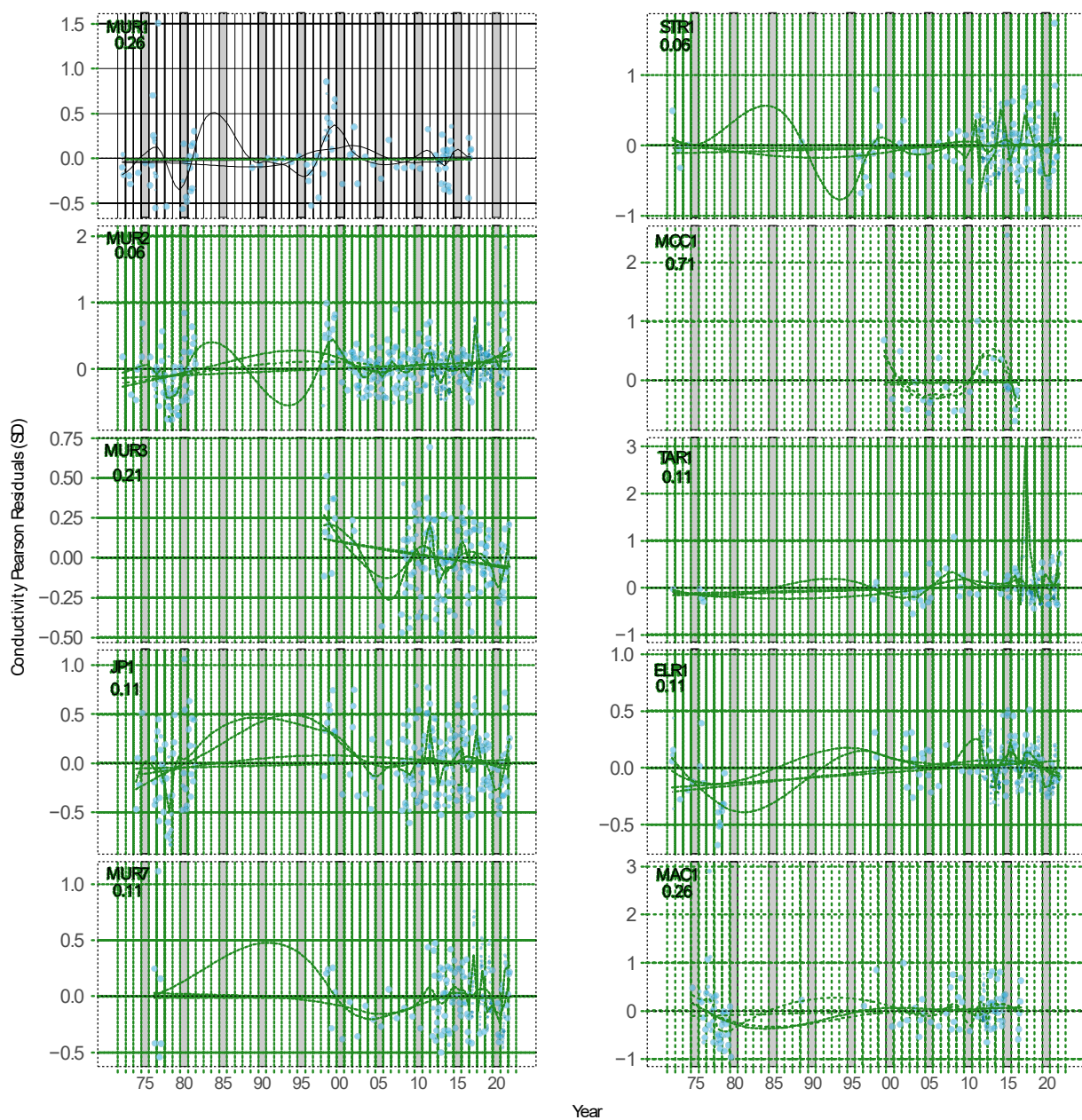


Figure S4 Pearson residuals for conductivity models calculated without Alkalinity (SD) in tributaries of the Oil Sands Region draining to the Athabasca River. Points are individual observations scaled by mean autocorrelation factors (Figure S8) and lines show smoothers (LOESS: 0.5 span in red; optimal span (values below site names) in black; GAM in green dashed; linear model in blue); Site names correspond to locations shown in Figure 1.

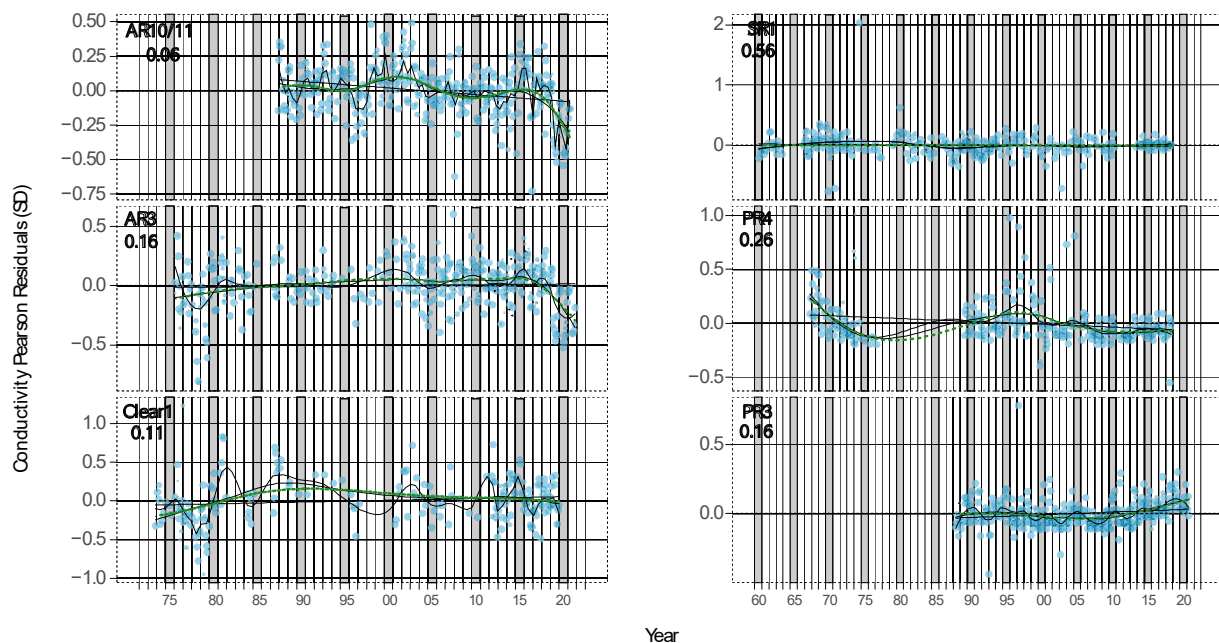


Figure S5 Pearson residuals for conductivity (SD) for large river locations for models calculated without alkalinity; Points are individual observations scaled by mean autocorrelation factors (Figure S8) and lines show smoothers (LOESS: 0.5 span in red; optimal span (values below site names) in black; GAM in green dashed; linear model in blue); Site names correspond to locations shown in Figure 1.

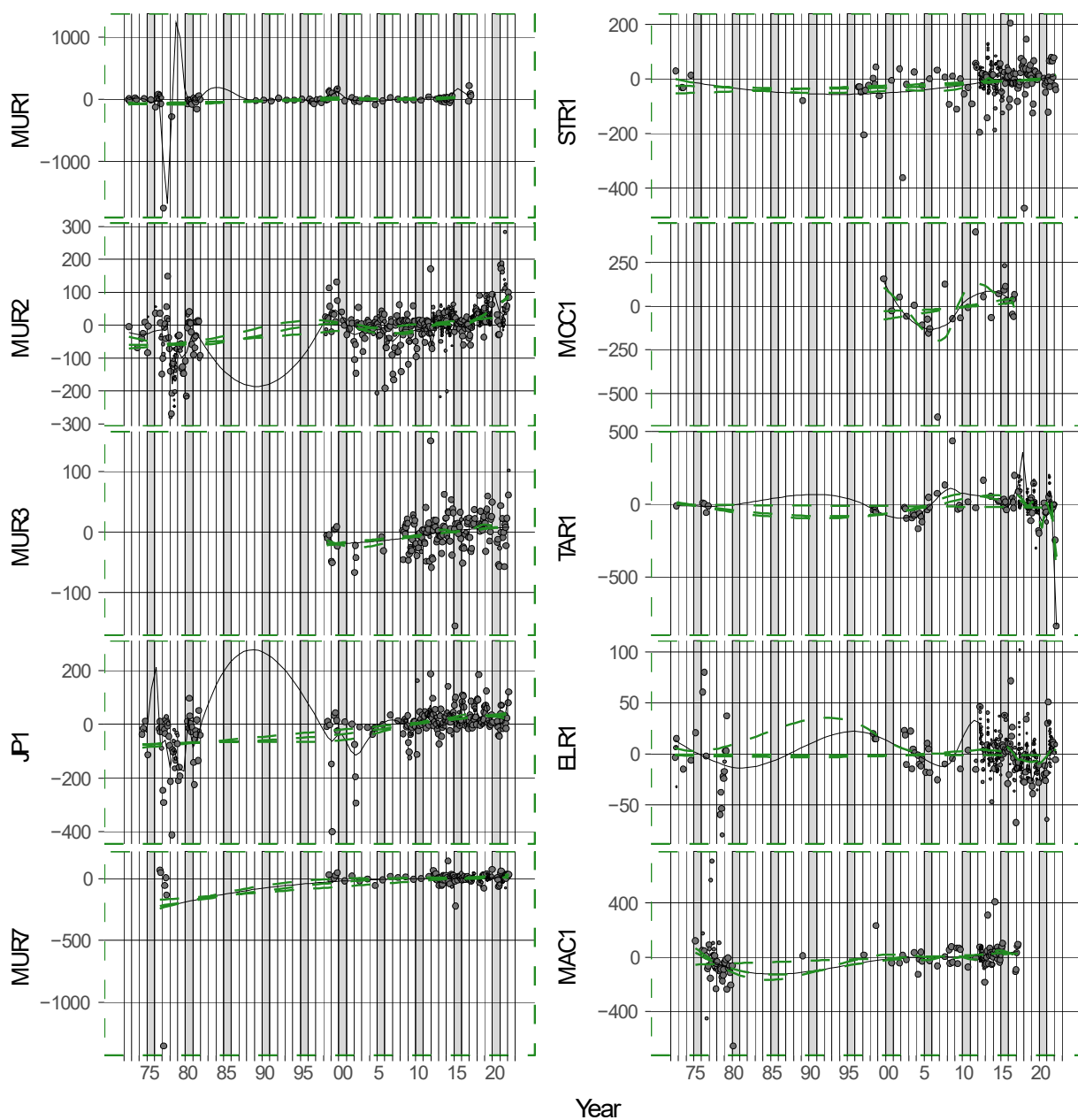


Figure S6 Response residuals for conductivity ($\mu\text{S}/\text{cm}$) in tributaries of the Oil Sands Region draining to the Athabasca River using models including alkalinity; points are individual observations scaled by mean autocorrelation factors (Figure S8) and lines show smoothers (LOESS: 0.5 span in red; optimal span (values below site names) in black; GAM in green dashed; linear model in blue); Site names correspond to locations shown in Figure 1.

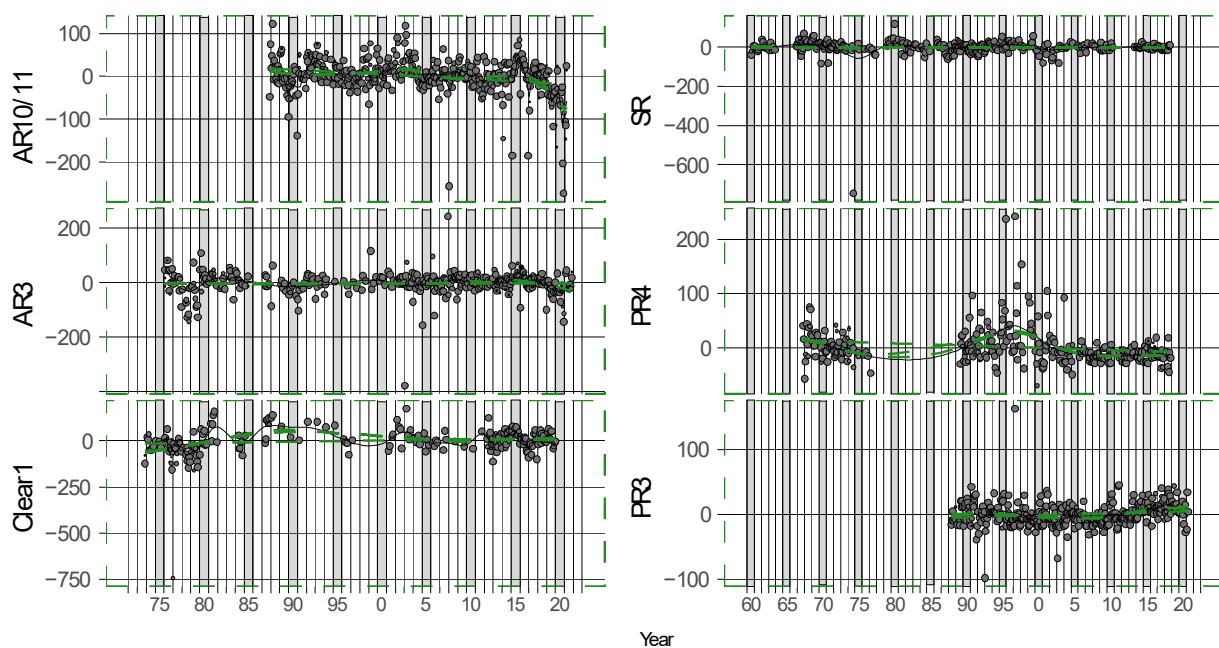


Figure S7 Response residuals for conductivity (uS/cm) in large river locations examined in this study using models including alkalinity; points are individual observations scaled by mean autocorrelation factors (Figure S8) and lines show smoothers (LOESS: 0.5 span in red; optimal span (values below site names) in black; GAM in green dashed; linear model in blue); Site names correspond to locations shown in Figure 1.

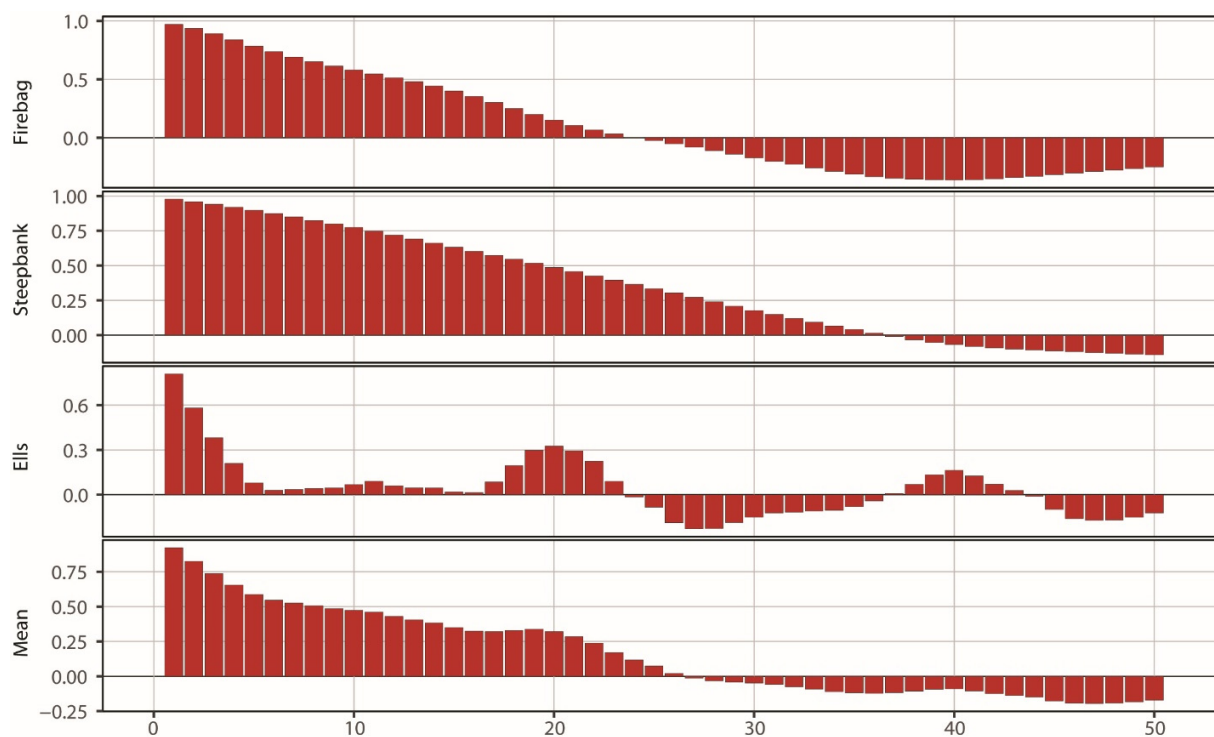


Figure S8 Autocorrelation coefficients for samples separated by 24-hr window from sondes deployed in 2013 in various tributaries; mean ACs used for weighting in smoothers; original data obtained from: <http://donnees.ec.gc.ca/data/substances/monitor/surface-water-quality-oil-sands-region/tributary-water-quality-oil-sands-region/>

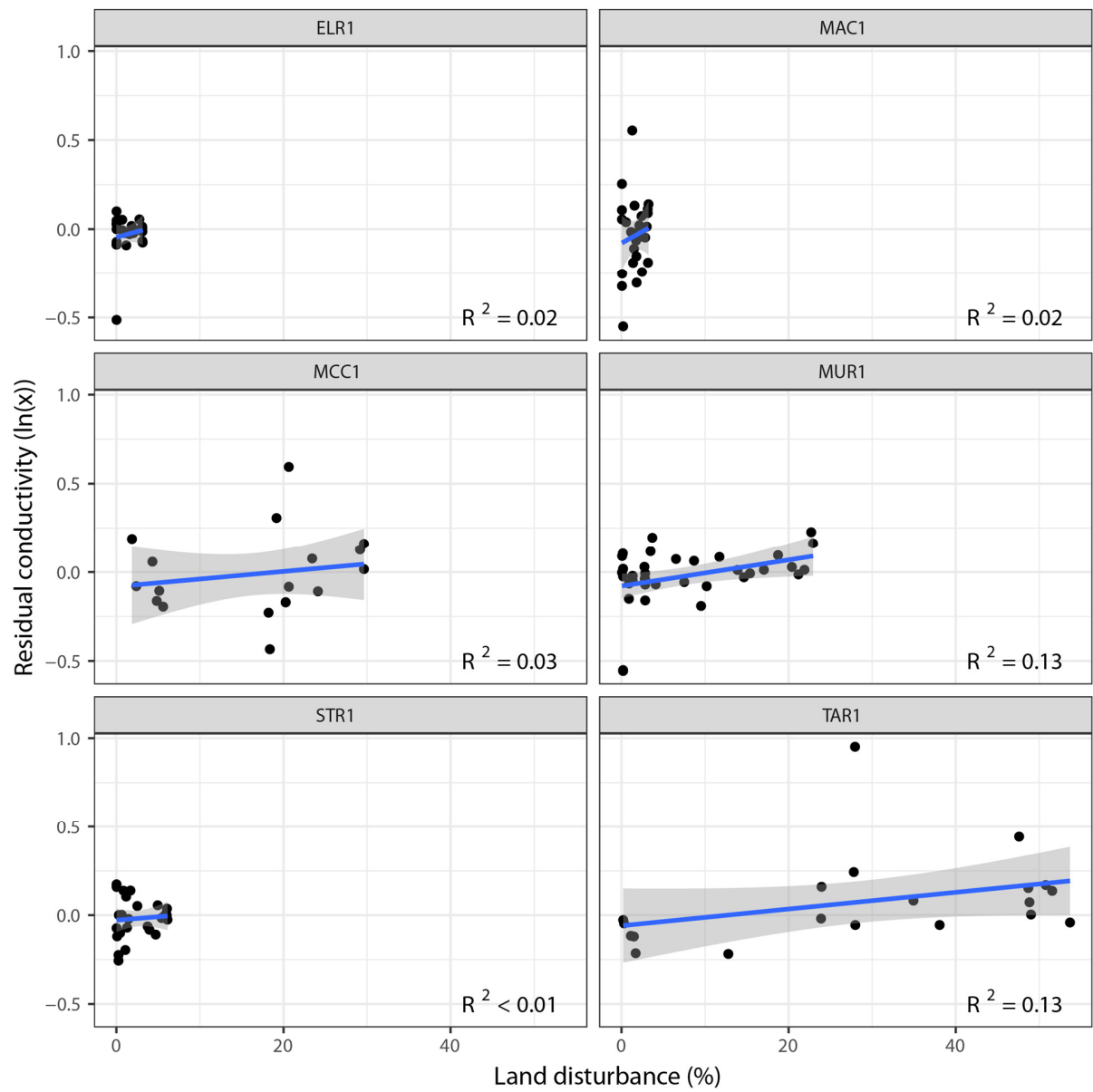


Figure S9 Residual conductivity compared to cumulative land disturbance at locations near mouths of the Athabasca tributaries in the AOSR.

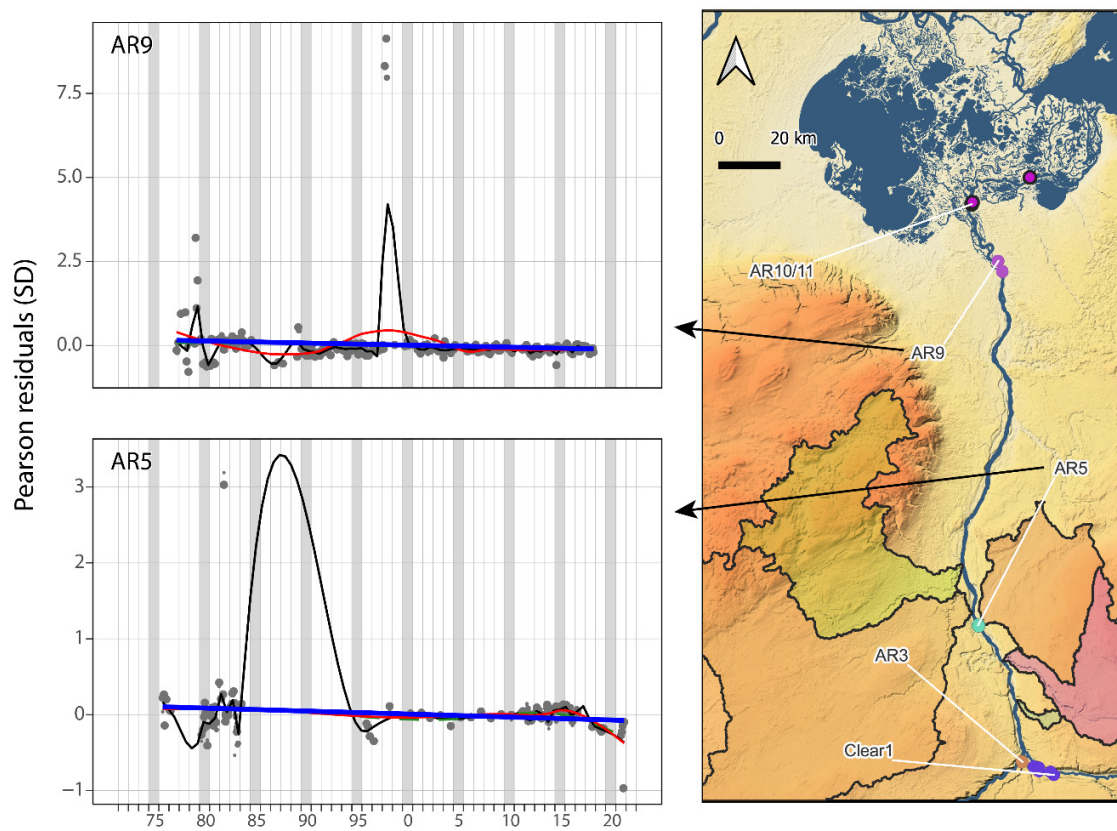


Figure S10 Additional residual conductivity data from AR5 and AR9.

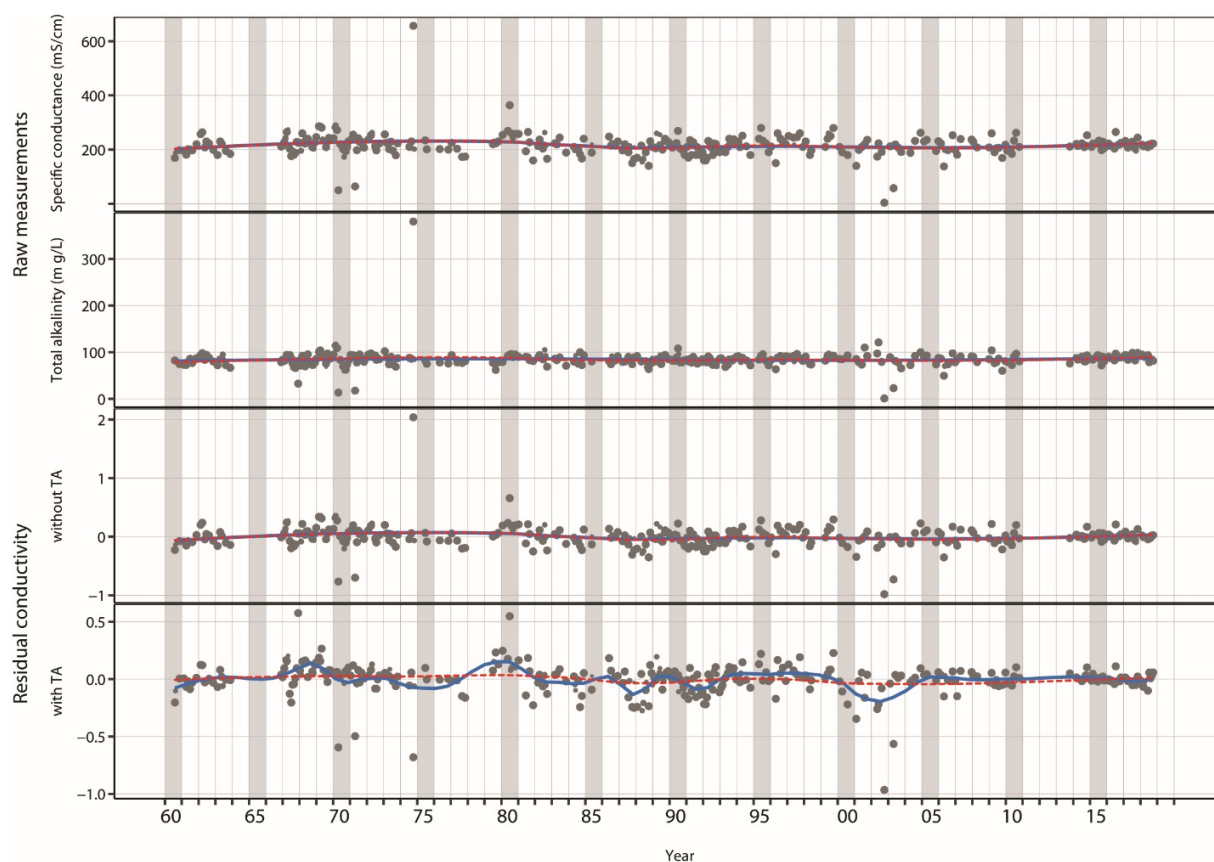


Figure S11 Comparisons of raw conductivity, raw Alkalinity, and residual conductivity (with and without total Alkalinity [TA]) at the SR1 location (Slave River at Fitzgerald) from 1960-2018; two smoothers shown: solid blue line = optimal span smoother; dashed red line = 0.5 span smoother.

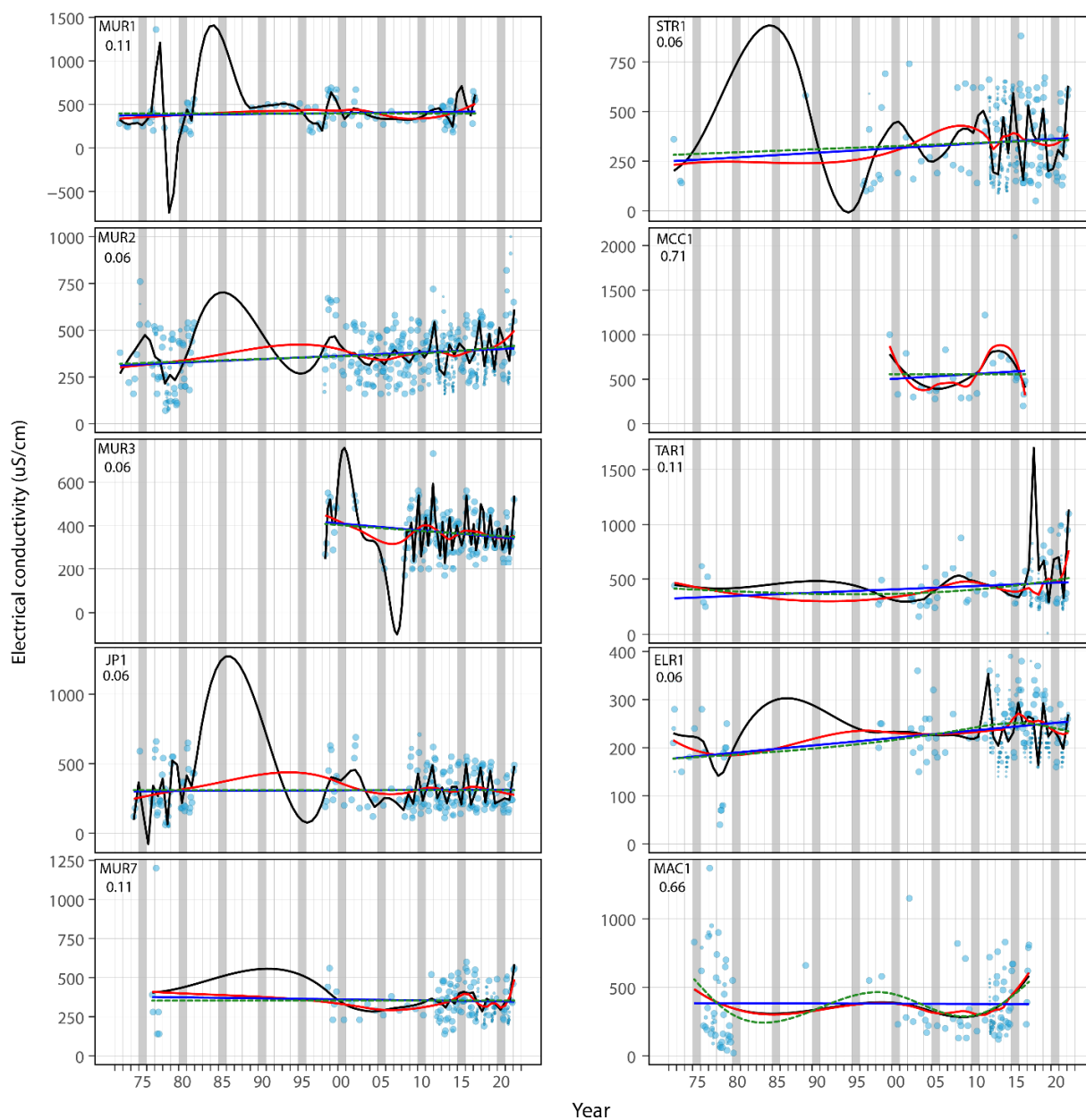


Figure S12 Conductivity (uS/cm) in tributaries of the Oil Sands Region draining to the Athabasca River. Points are individual observations scaled by mean autocorrelation factors (Figure S8) and lines show smoothers (LOESS: 0.5 span in red; optimal span (values below site names) in black; GAM in green dashed; linear model in blue); Site names correspond to locations shown in Figure 1.

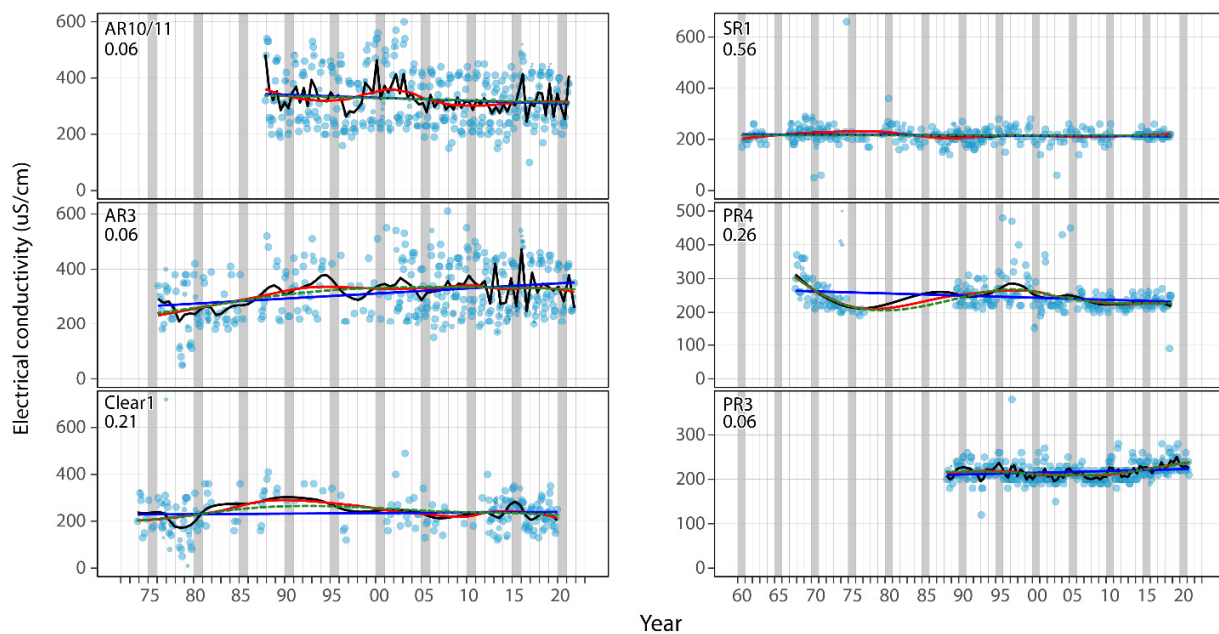


Figure S13 Conductivity (uS/cm) in large river locations examined in this study. Points are individual observations scaled by mean autocorrelation factors (Figure S8) and lines show smoothers (LOESS: 0.5 span in red; optimal span (values below site names) in black; GAM in green dashed; linear model in blue); Site names correspond to locations shown in Figure 1.