



The spatio-temporal response of summertime tropospheric ozone to changes in precursor emissions in the Lower Fraser Valley, British Columbia

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A Retrospective Analysis of Ozone Formation in the Lower Fraser Valley, Canada.

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Presented to the NW-Airquest annual meeting Pullman Wa., June 8, 2012.

Follow-up study to work
presented here in 2012

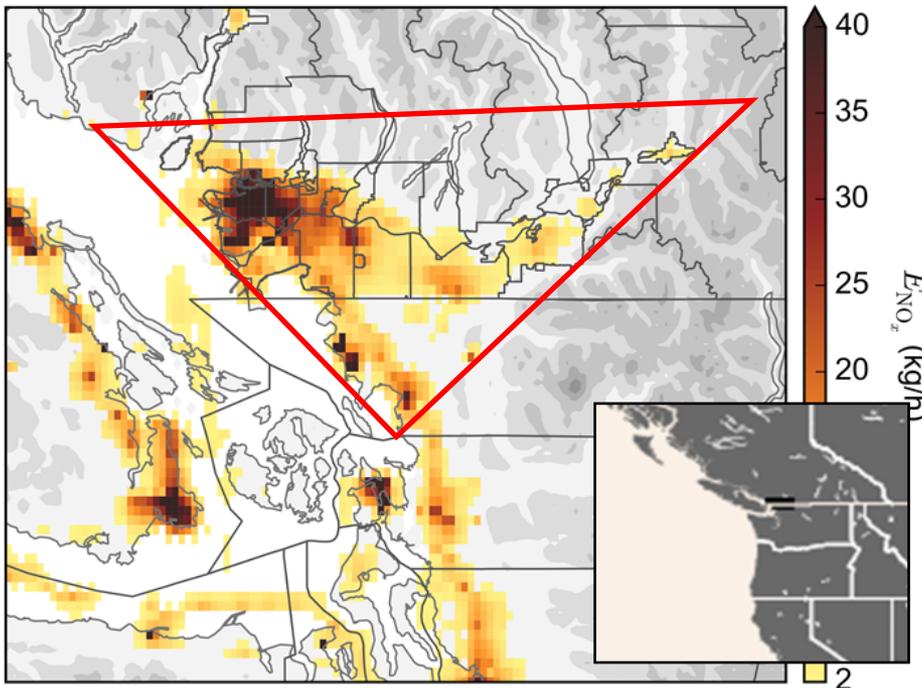
- That was a modeling study
- This is a statistical data-driven study

Then, as now, the research question is:

How have summertime O_3 concentrations changed in the LFV over the last 26 years, given large scale changes in local emissions?

Explored through an examination of:

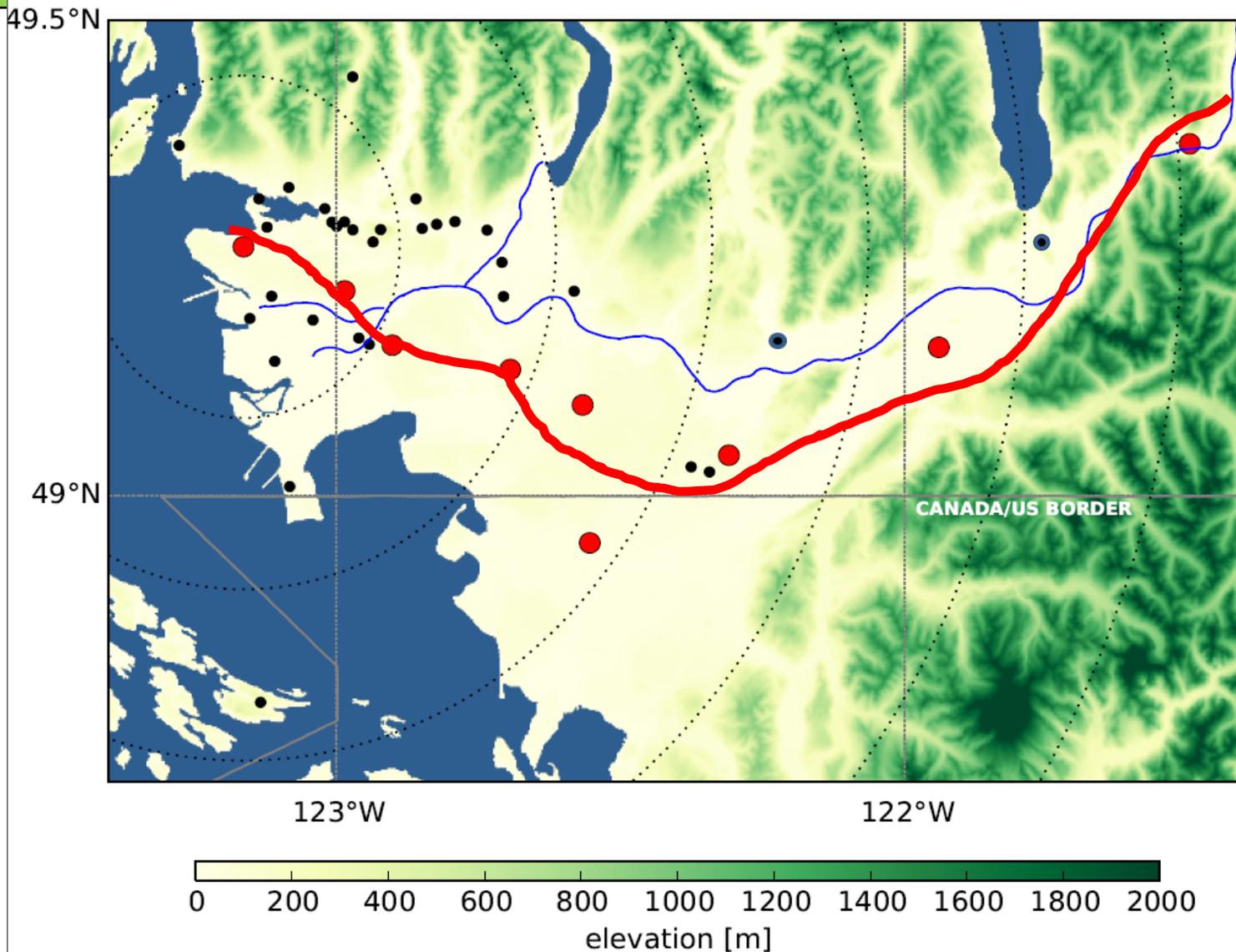
1. Trends in emissions
2. Trends in summer $[NO_x]$, $[VOC]$ at key stations
3. Trends in summer $[O_3]$ at different levels at key stations



LFV

- Triangular coastal valley
- ~2 million people
- Straddles US-Canada border

Spatio-temporal analysis using monitoring data along an east-west transect of stations



Statistical Models:

Daily summertime (JJA) data from 9 monitors across 26 years (1990-2015) [Potentially $92 \times 26 \times 9 = 21528$ data points]

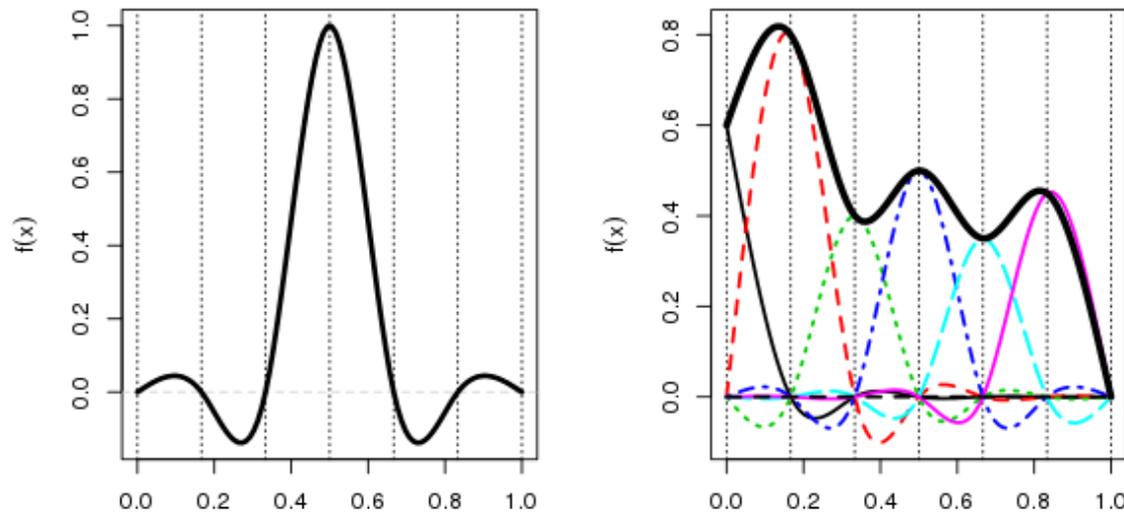
Pollutant	Description
NOx	Daytime (0600-1800) average
VOC	24-hour average
Ozone - Background	10 th percentile and below of lowest variance MD8A
Ozone – Fair weather	Sunny fair weather (roughly 50-96 th percentile) MD8A
Ozone – CAAQS	Summer time four highest MD8A
Others	fraction of OH reacting with NO ₂

General Additive Models (GAM): *response of variable to predictor is not always linear*

Fit Y using some unknown but potentially non-linear function of X

$$y_i = f(x_i) + \epsilon_i,$$

How to choose f? – Use basis of spline curves

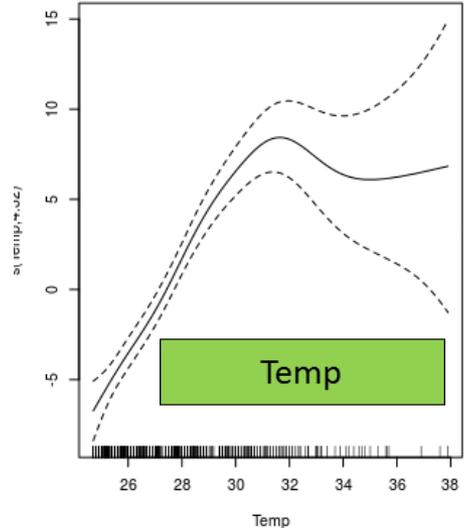


<http://www.fromthebottomoftheheap.net/2014/05/09/modelling-seasonal-data-with-gam/>

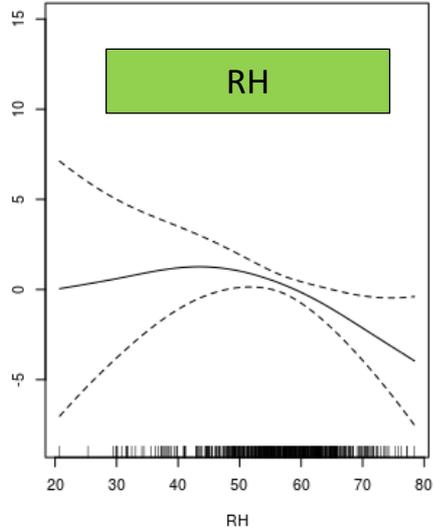
$$f(x) = \sum_{i=1}^q b_i(x)\beta_i,$$

$$[O_3] \sim s(Temp) + s(RH) + s(DoY) + s(Ucpt, Vcpt) + s(Year) + \epsilon$$

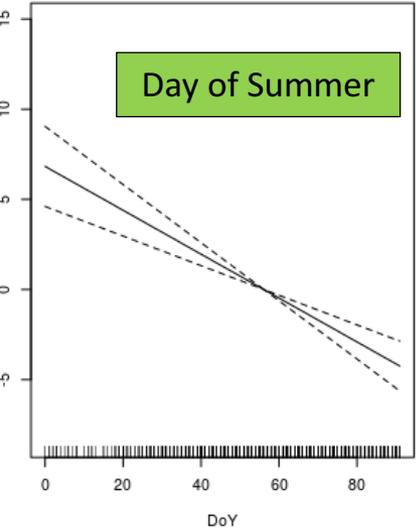
$[O_3] = \text{Const} +$



+

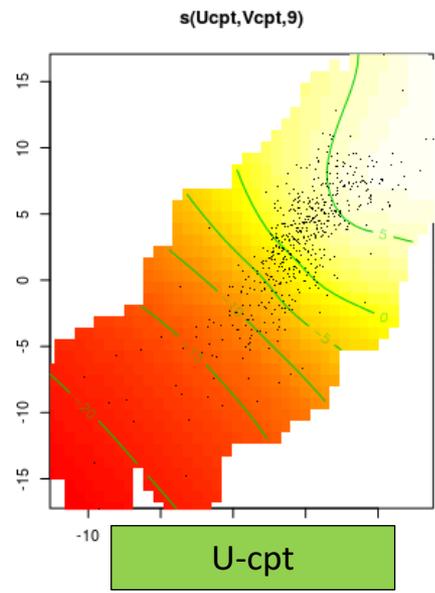


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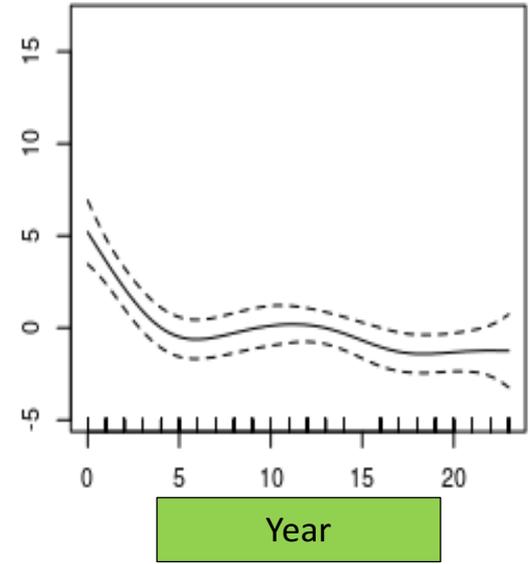


+

V-cpt



+



+ ϵ

Typical Model formulation

Pollutant

Cultural effects (e.g. weekend/weekday effect)

Long term network response



Local station temporal response

Canonical Footprint



Meteorological effects



$$[O_3] \sim s(Temp, RH) \times s(ETot, DoY) \times s(Ucpt, Vcpt) \times s(WeekDay) \times s(Dist) \times s(Year) \times ti(Dist, Year) + \epsilon$$

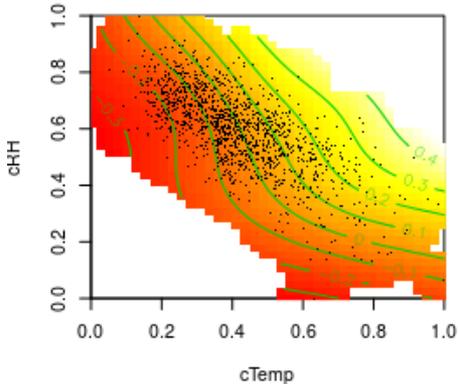
Variable	Description
Temp	Daily maximum temperature
RH	Daytime average RH
ETot	Daytime total incoming SW radiation from T09
DoY	JJA day number (1-92)
Ucpt, Vcpt	Average daytime wind cpts from YVR or YXX
Weekday	Day of week
Dist	Distance from T02
Year	1990 to 2015

We are after the yellow boxes - the other factors used to isolate the trend

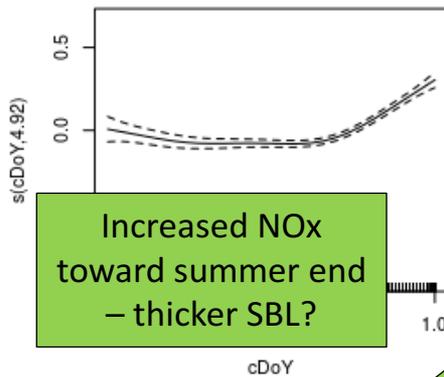


NOx Results

te(cTemp,cRH,9.17)

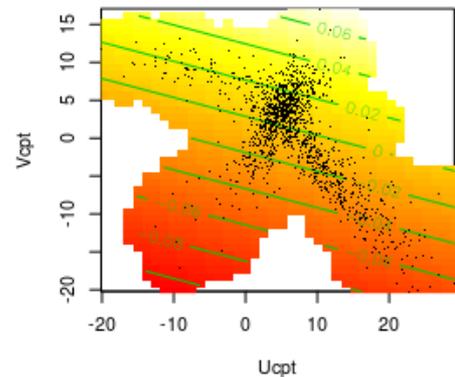
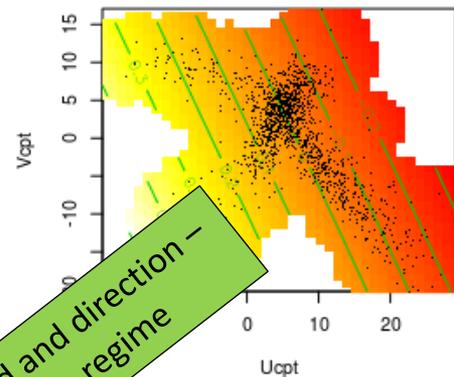


s(Ucpt,Vcpt,1.97):Meso_f1



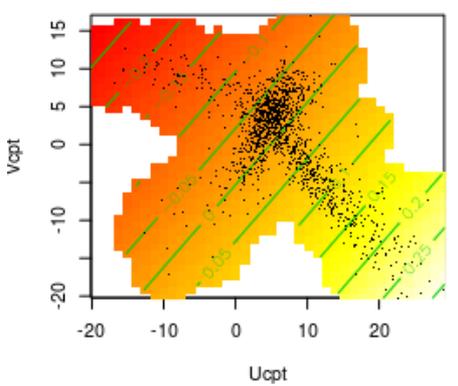
Increased NOx
toward summer end
– thicker SBL?

s(Ucpt,Vcpt,1.51):Meso_f2

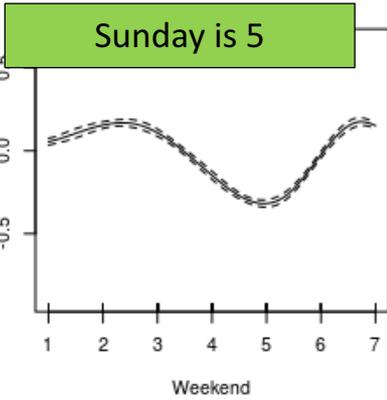
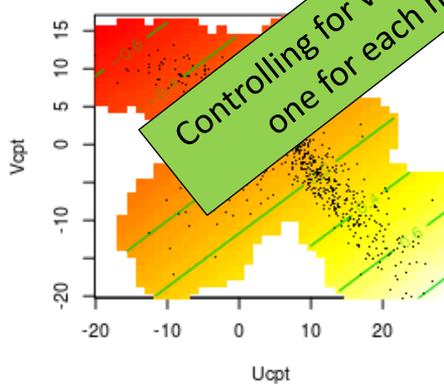


Controlling for wind speed and direction –
one for each meso-scale regime

s(Ucpt,Vcpt,1.66):Meso_f3

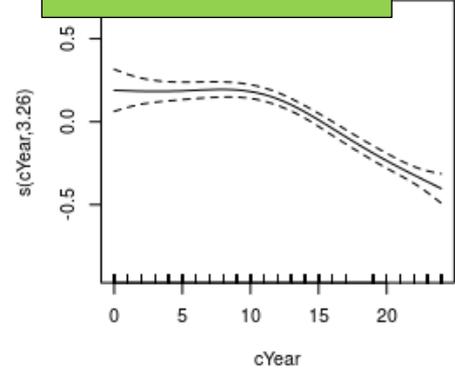


s(Ucpt,Vcpt,2.02):Meso_f4

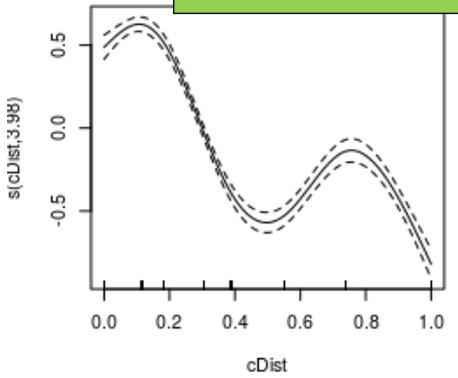


Sunday is 5

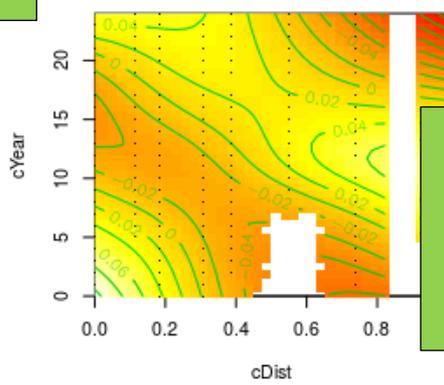
Network-wide trend



NOx footprint



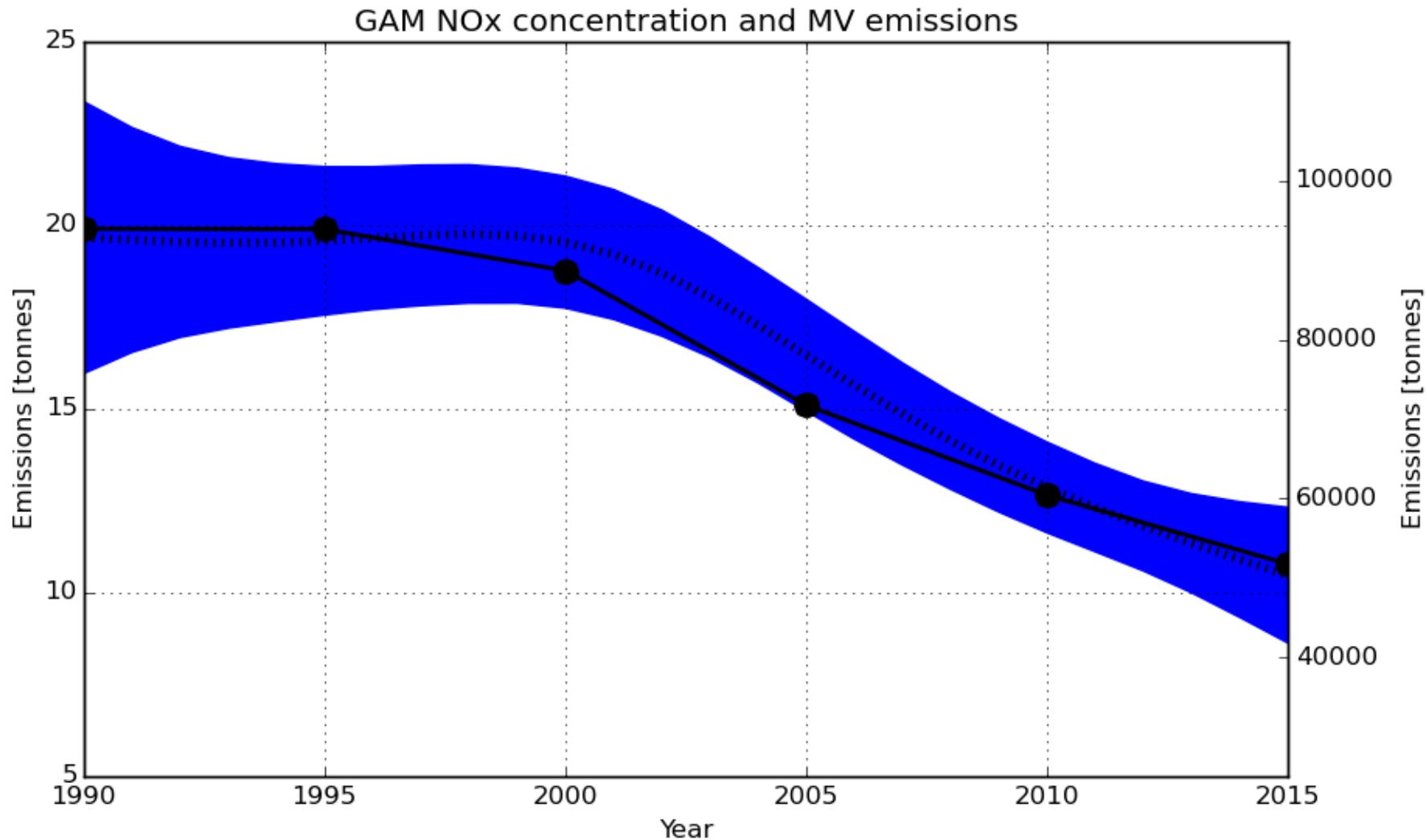
tl(cDist,cYear,4.56)



Local changes in NOx over
time – above and beyond
canonical footprint and
network-wide trend – not
uniform!

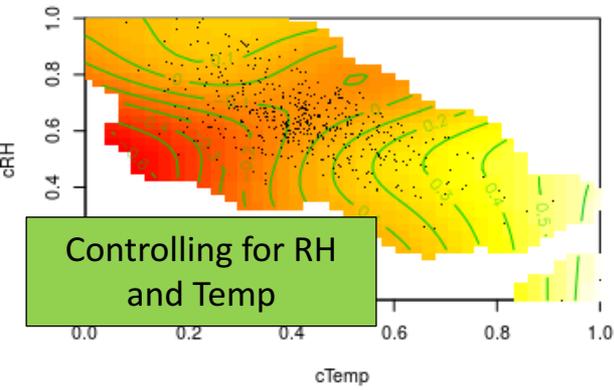
GAM NOx
Smooths

GAM-predicted networked averaged NOx versus MV emissions totals

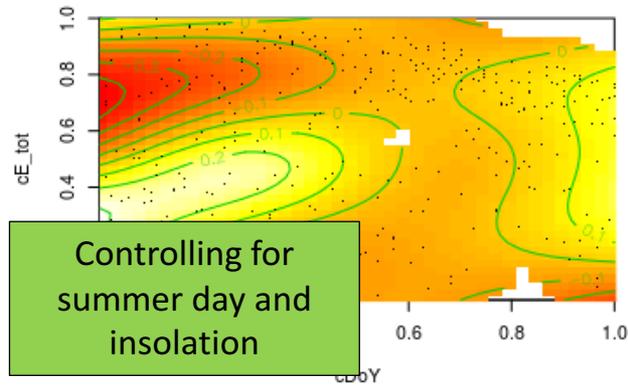


VOC Results

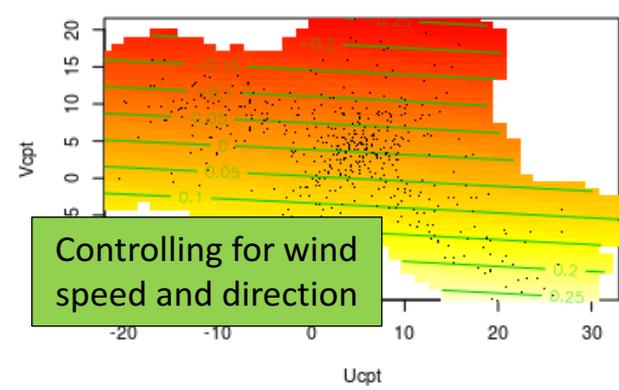
te(cTemp,cRH,7.87)



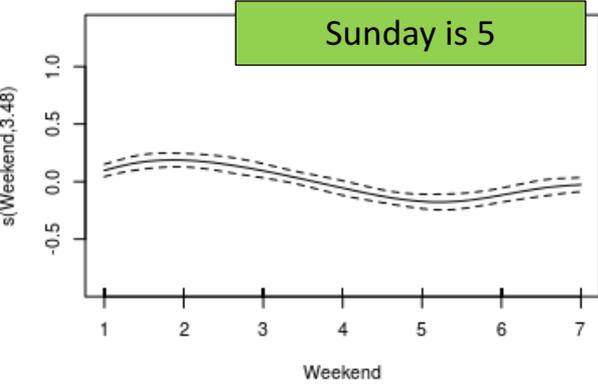
te(cDoY,cE_tot,13.31)



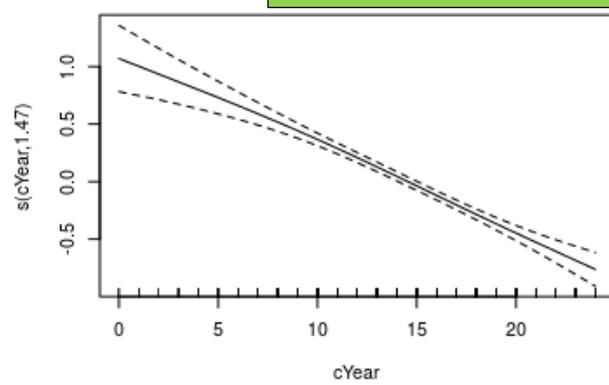
s(Ucpt,Vcpt,1.8)



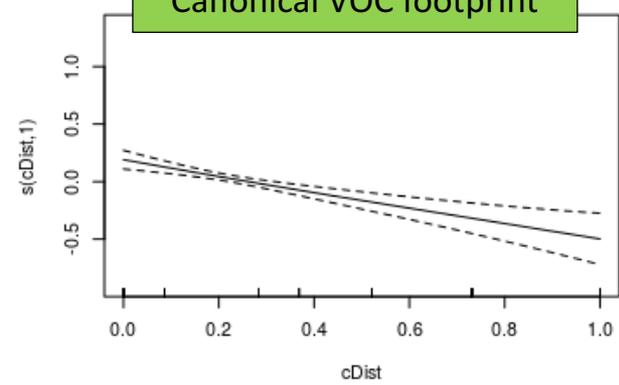
Sunday is 5



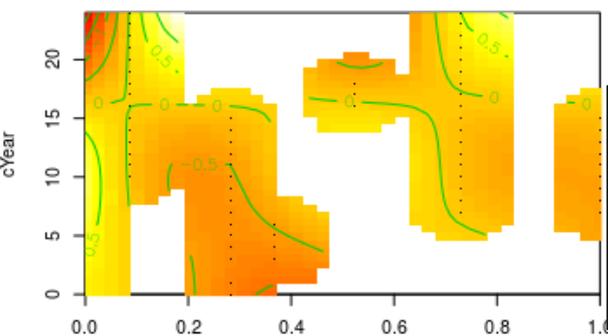
Network-wide trend



Canonical VOC footprint

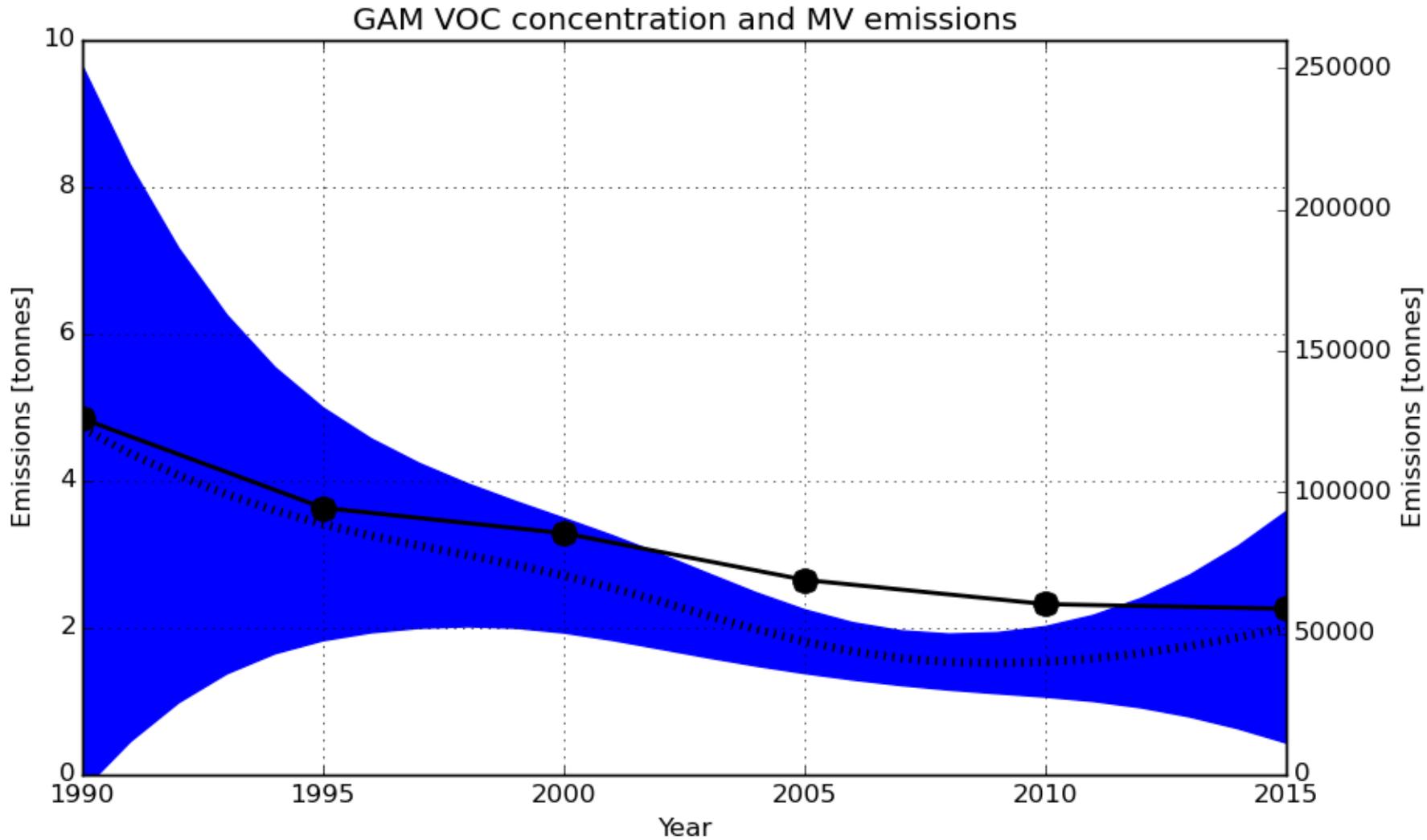


ti(cDist,cYear,9.8)



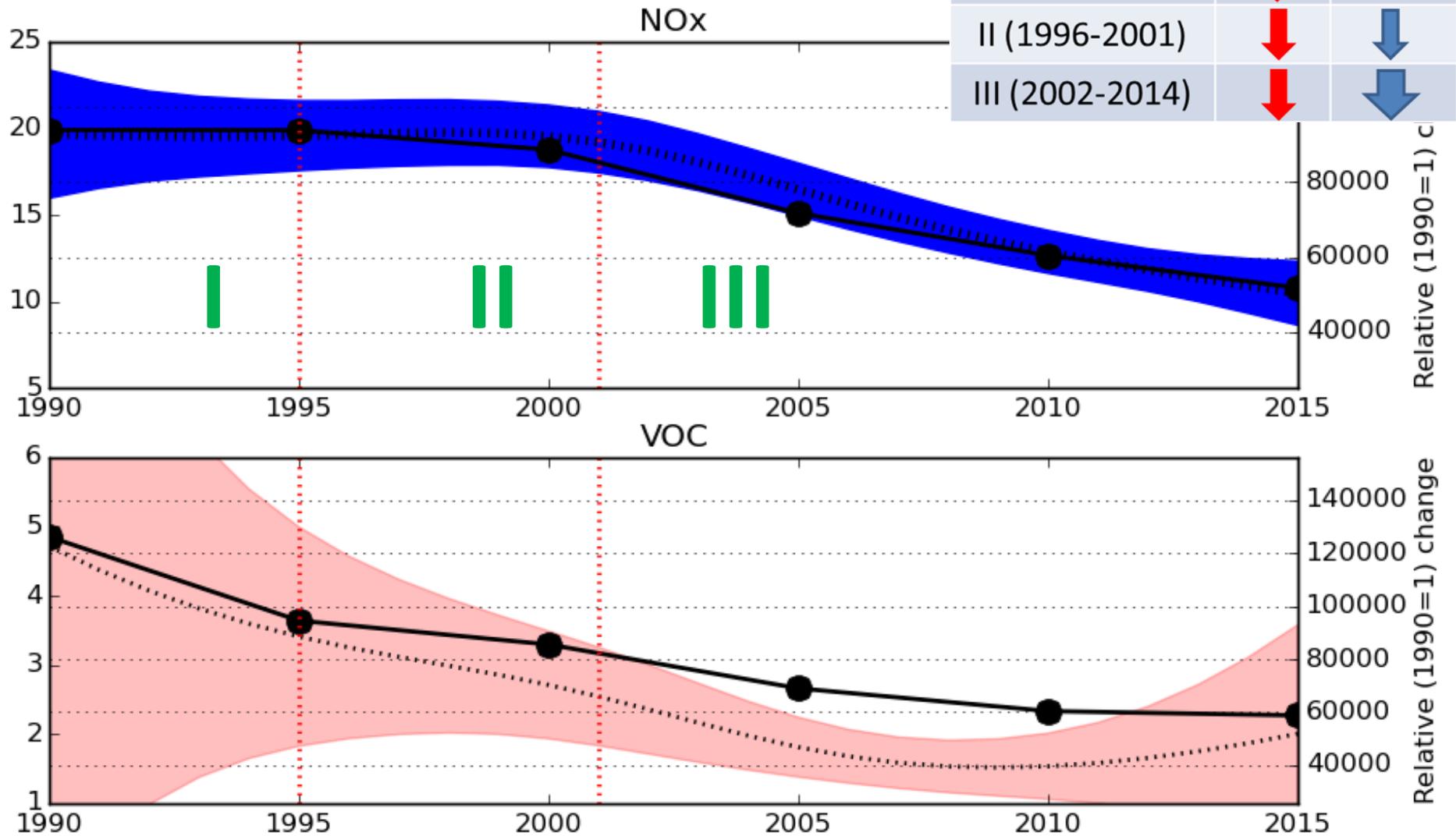
GAM VOC Smooths

GAM-predicted networked averaged VOC versus MV emissions totals



Era	EVOC	ENox
I (1990-1995)		
II (1996-2001)		
III (2002-2014)		

GAM annual conc. trends (shaded area) and MV annual emissio



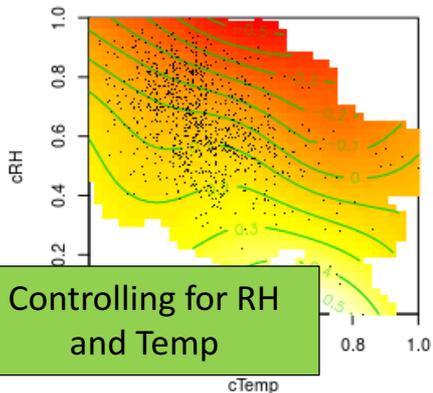
Precursor Summary

- GAM annual average concentration trends consistent with MV annual emissions trends
- VOC results are more uncertain – likely due to limited sampling on the GAM-side and difficulty quantifying fugitive and evaporative emissions on the MV-side
- VOC and NOx annual trends are not linear
- Rate of VOC reductions (emissions or concentrations) have **not** occurred at the same rate as NOx reductions

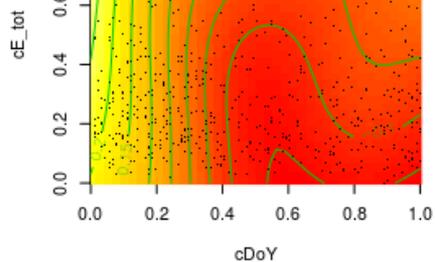
Ozone Results

Background days

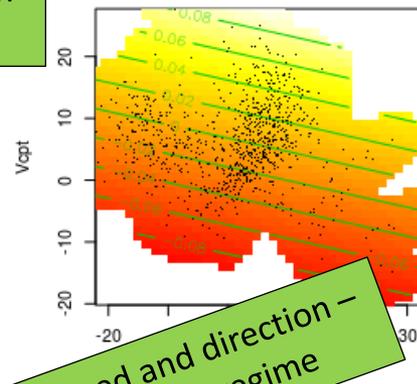
te(cTemp,cRH,10.86)



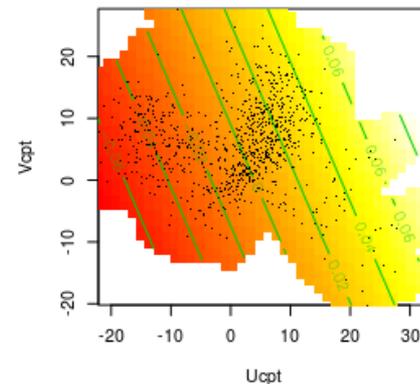
Controlling for insolation and summer day



s(Ucpt,Vcpt,1.5):NCEP_f0

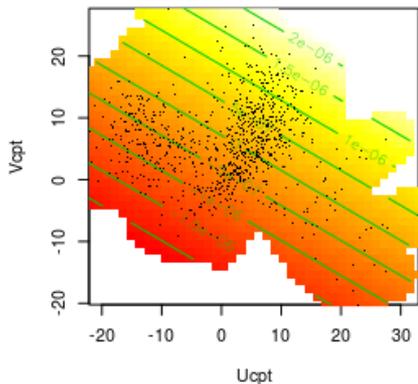


s(Ucpt,Vcpt,1.68):NCEP_f1

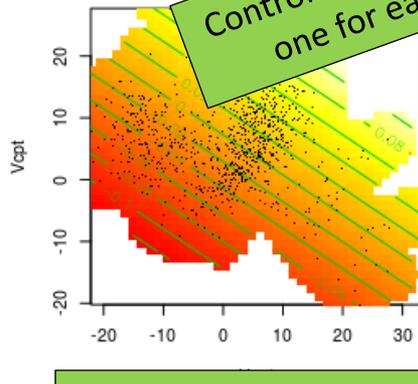


Controlling for wind speed and direction - one for each synoptic-scale regime

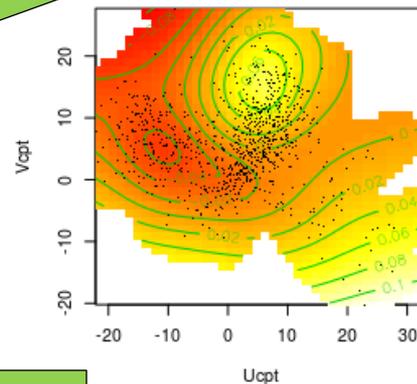
s(Ucpt,Vcpt,0):NCEP_f2



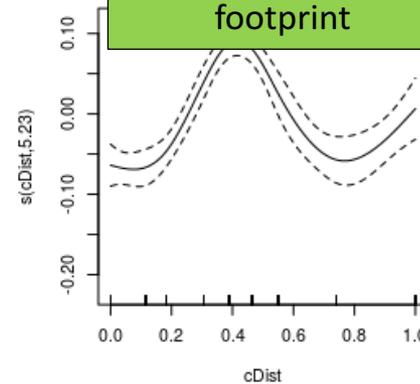
s(Ucpt,Vcpt,1.68):NCEP_f3



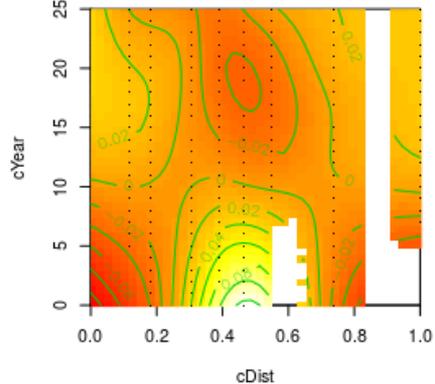
s(Ucpt,Vcpt,6.63):NCEP_f4



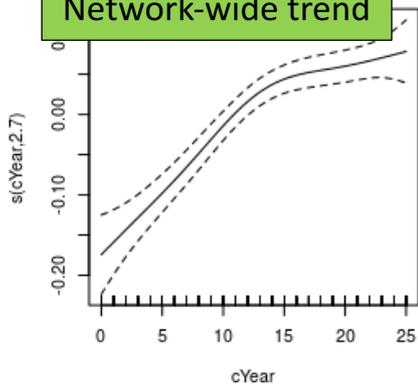
Canonical ozone footprint



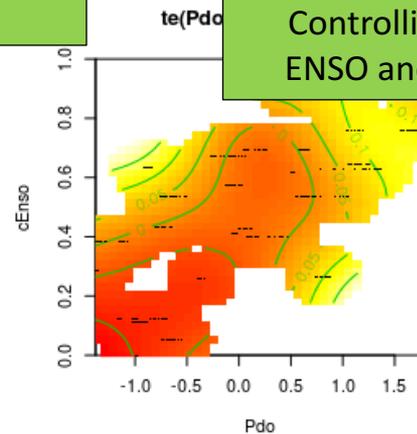
Local changes in ozone- not uniform!



Network-wide trend



Controlling for ENSO and PDO



Have all monitors responded equally to
changing background?
(after accounting for location and meteorology)

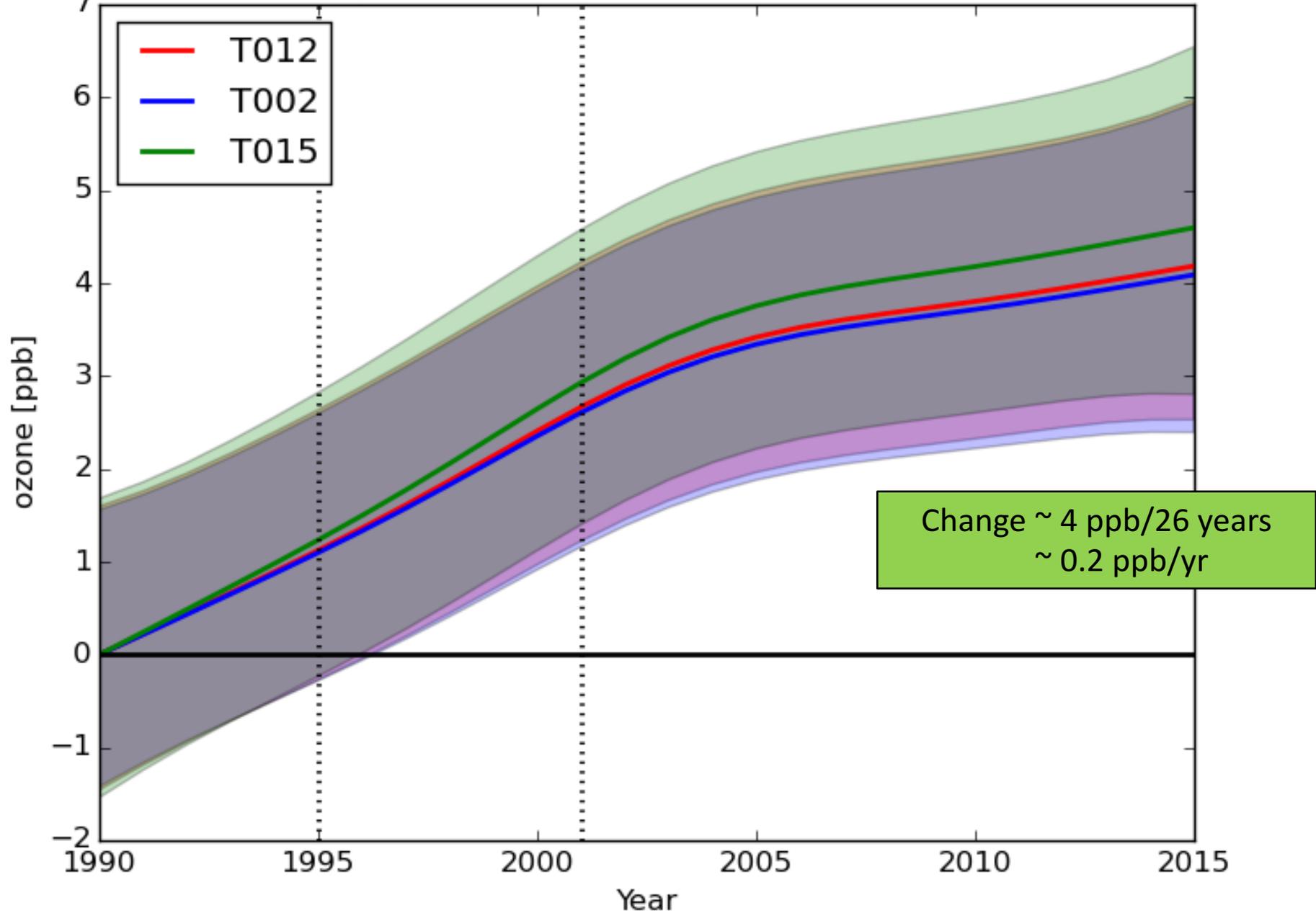
$$H_0 : [O_3] \sim s(Temp, RH) \times s(ETot, DoY) \times s(Ucpt, Vcpt) \times s(WeekDay) \times s(Dist) \times s(Year) \times ti(Dist, Year) + \epsilon$$

$$H_1 : [O_3] \sim s(Temp, RH) \times s(ETot, DoY) \times s(Ucpt, Vcpt) \times s(WeekDay) \times s(Dist) \times s(Year) + \epsilon$$

Model	df	BIC	AIC
H0	19	-348	-452
H1	16	-363	-450

Hard to say!
BIC score for model without
Dist+Year interaction (H_1) is
more negative
But AIC slight favours H_0

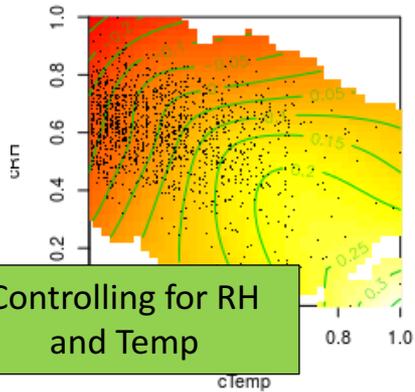
Relative change (1990=0) in ozone BACKGROUND over time



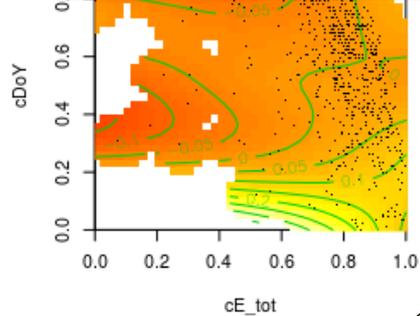
Ozone Results

Fair weather days

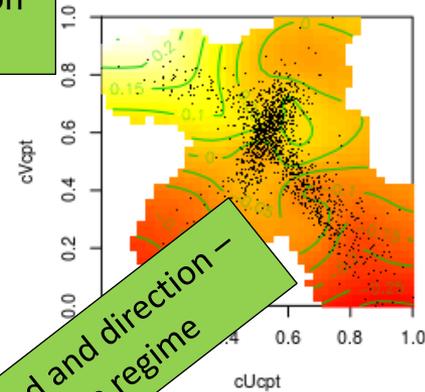
te(cTemp,cRH,13.36)



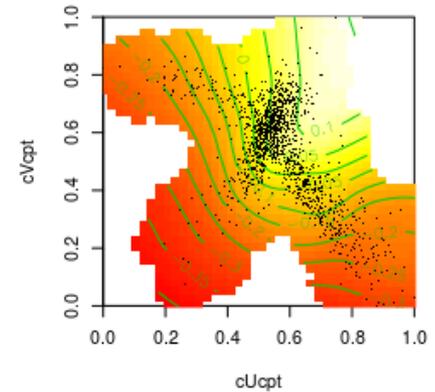
Controlling for insolation and summer day



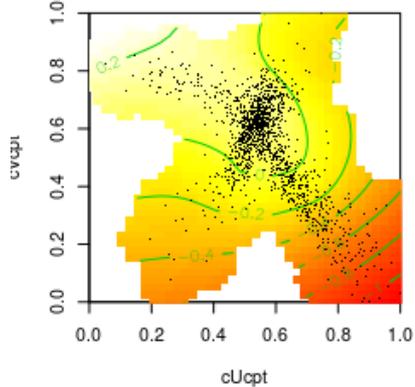
s(cUcpt,cVcpt,17.18):Meso_f1



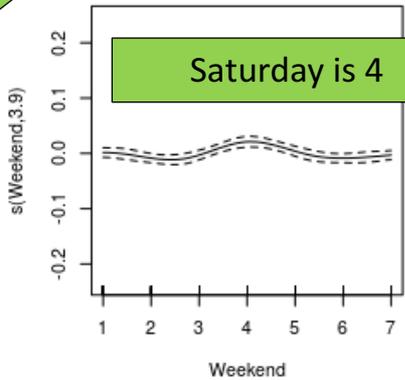
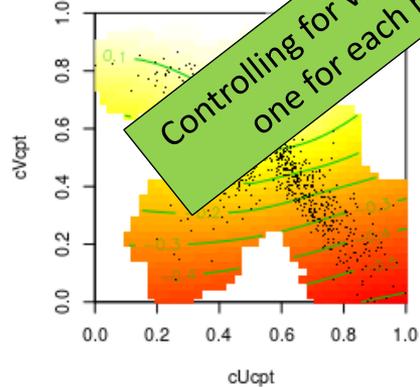
s(cUcpt,cVcpt,11.5):Meso_f2



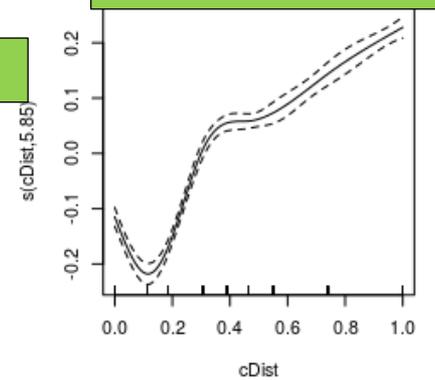
s(cUcpt,cVcpt,12.59):Meso_f3



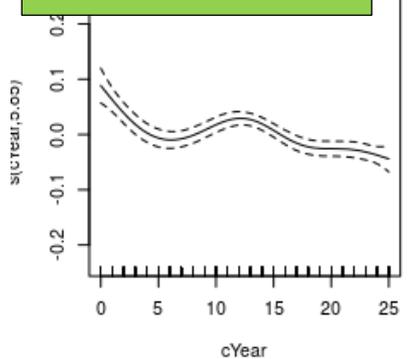
s(cUcpt,cVcpt,8.38):M



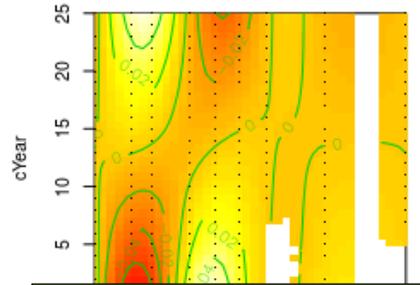
Canonical ozone footprint



Network-wide trend



tl(cDist,cYear,5.8)



Local changes in ozone - not uniform!

GAM Ozone - 'fair weather days' Smooths

Have all monitors responded equally to
changing emissions?
(after accounting for location and meteorology)

$$H_0 : [O_3] \sim s(Temp, RH) \times s(ETot, DoY) \times s(Ucpt, Vcpt) \times s(WeekDay) \times s(Dist) \times s(Year) \times ti(Dist, Year) + \epsilon$$

$$H_1 : [O_3] \sim s(Temp, RH) \times s(ETot, DoY) \times s(Ucpt, Vcpt) \times s(WeekDay) \times s(Dist) \times s(Year) + \epsilon$$

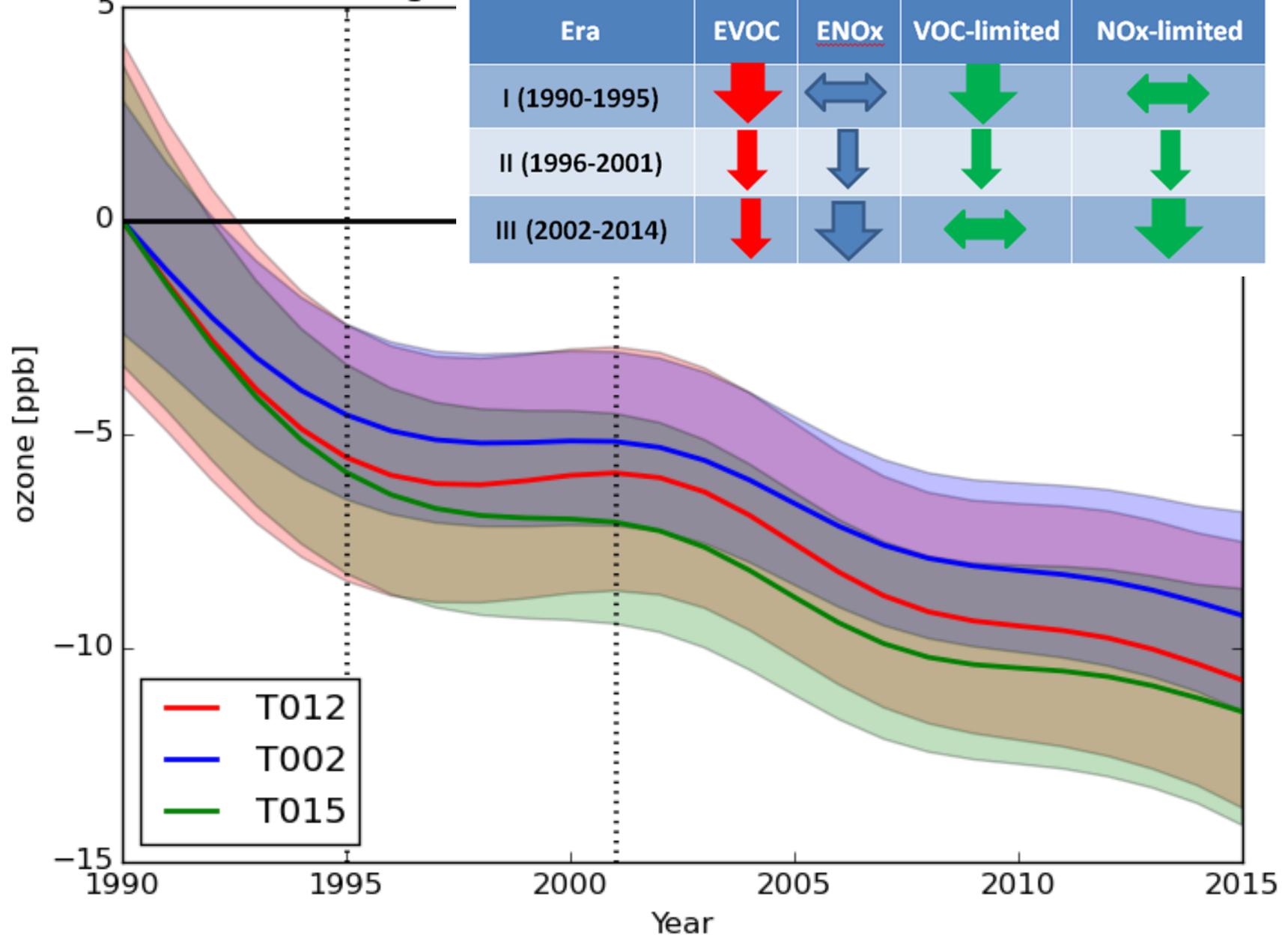
Model	df	BIC	AIC
H0	33	-3516	-3733
H1	31	-3506	-3710

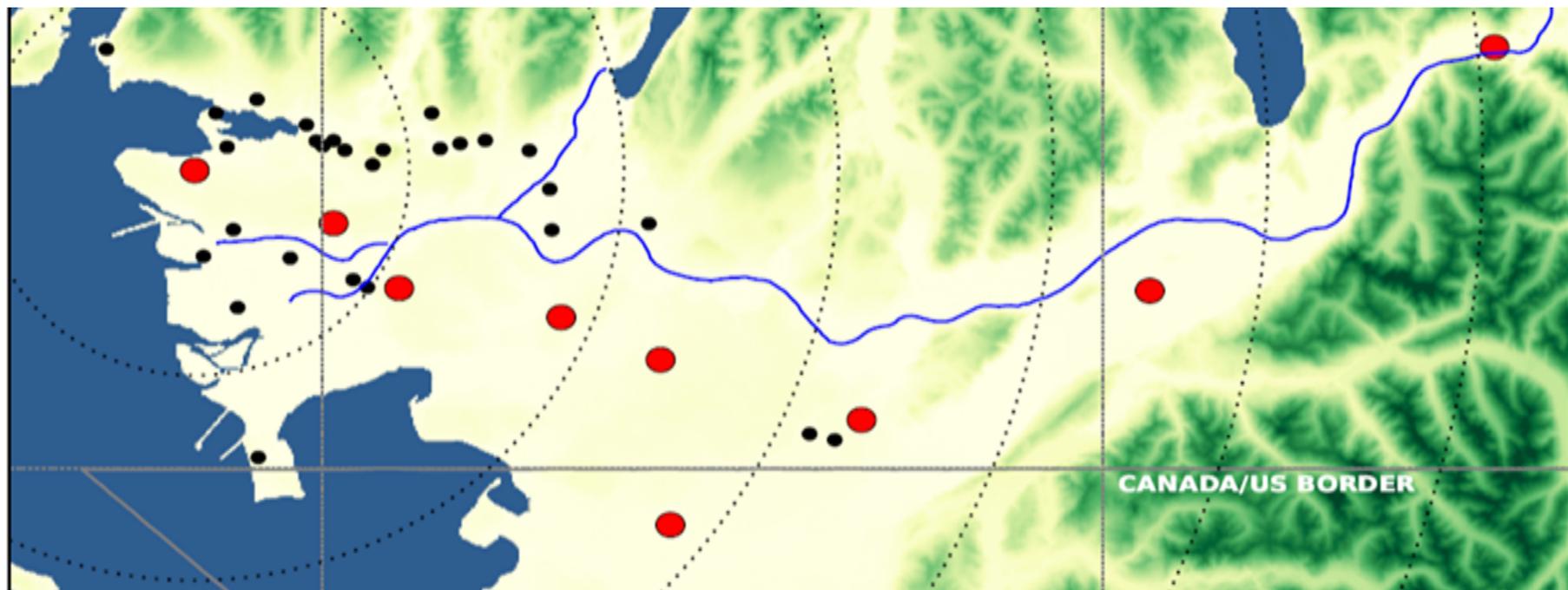
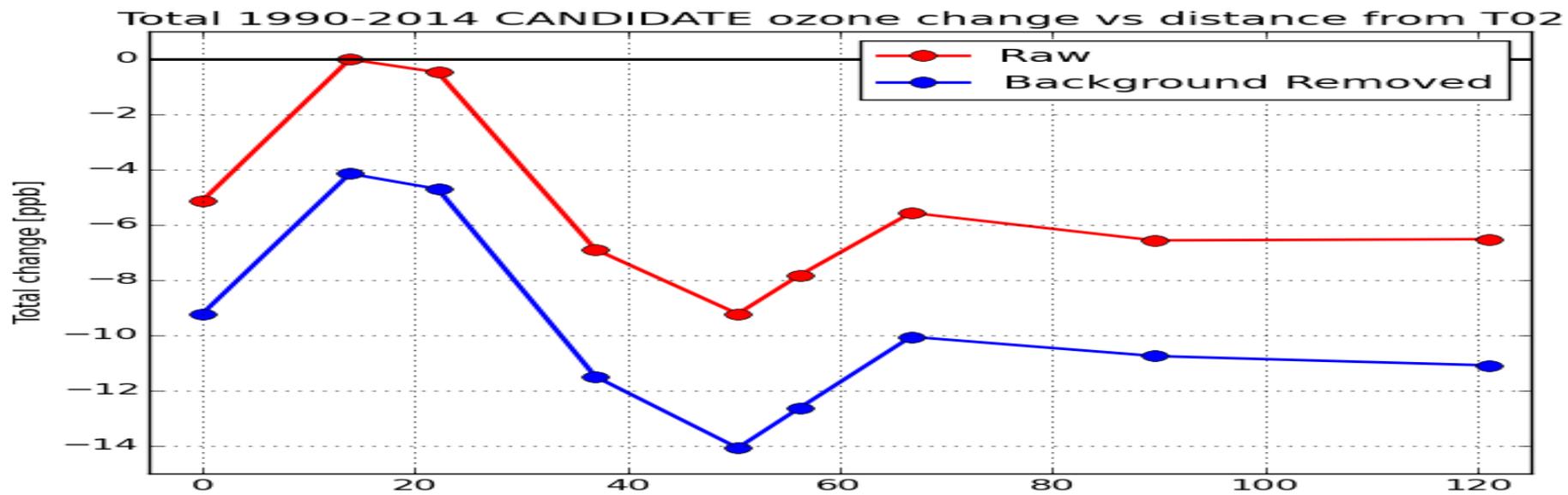
NO!

Both BIC and AIC scores support (are more negative) the H₀ model with Dist+Year interaction term.

Relative change (1990-0) in ozone CANDIDATE over time

Era	EVOC	ENOx	VOC-limited	NOx-limited
I (1990-1995)	↓	↔	↓	↔
II (1996-2001)	↓	↓	↓	↓
III (2002-2014)	↓	↓	↔	↓

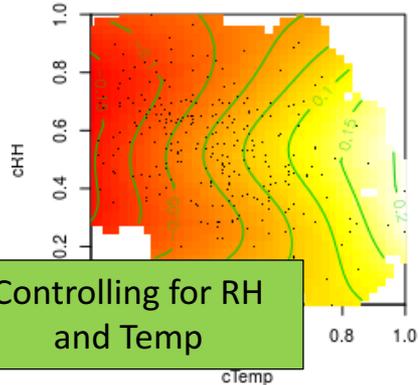




Ozone Results

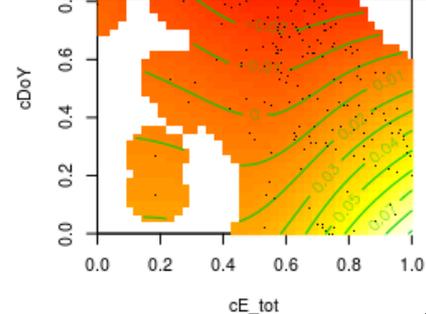
CAAQS days

te(cTemp,cRH,8.54)

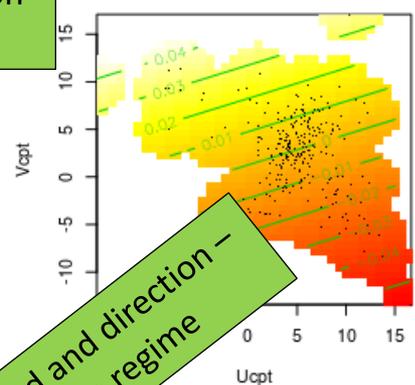


Controlling for RH and Temp

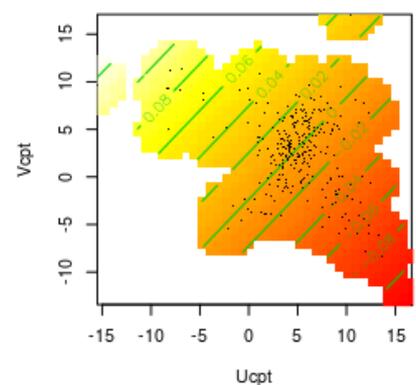
Controlling for insolation and summer day



s(Ucpt,Vcpt,1.15):Meso_f1

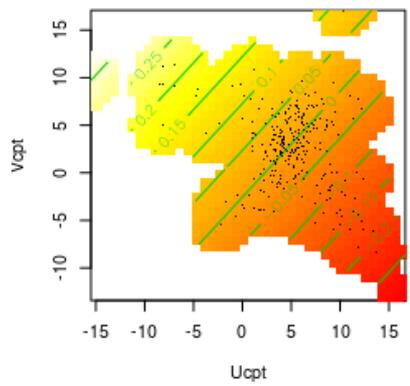


s(Ucpt,Vcpt,1.56):Meso_f2

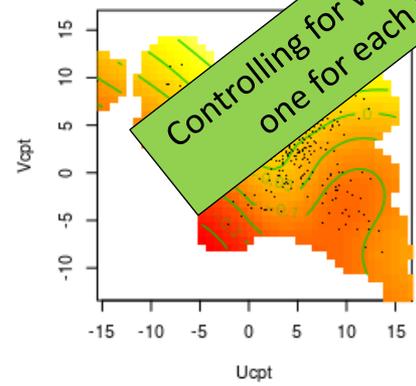


Controlling for wind speed and direction – one for each meso-scale regime

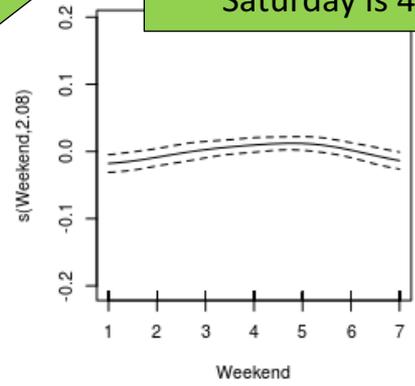
s(Ucpt,Vcpt,1.85):Meso_f3



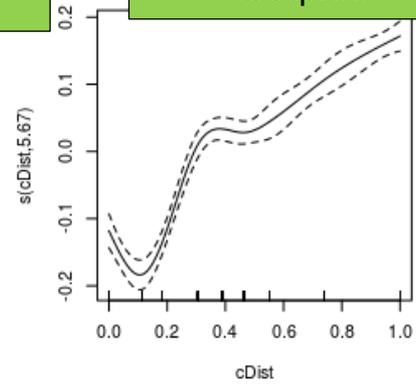
s(Ucpt,Vcpt,7.22):Meso_f4



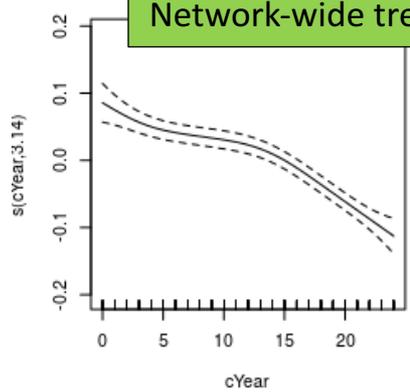
Saturday is 4



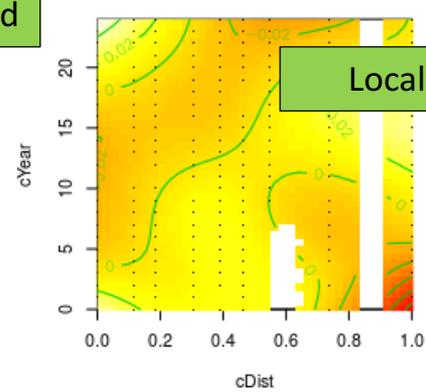
Canonical ozone footprint



Network-wide trend



ti(cDist,cYear,8.65)



Local changes in ozone

GAM Ozone – 'CAAQO days' Smooths

Have all monitors responded equally to
changing emissions?
(after accounting for location and meteorology)

$$H_0 : [O_3] \sim s(Temp, RH) \times s(ETot, DoY) \times s(Ucpt, Vcpt) \times s(WeekDay) \times s(Dist) \times s(Year) \times ti(Dist, Year) + \epsilon$$

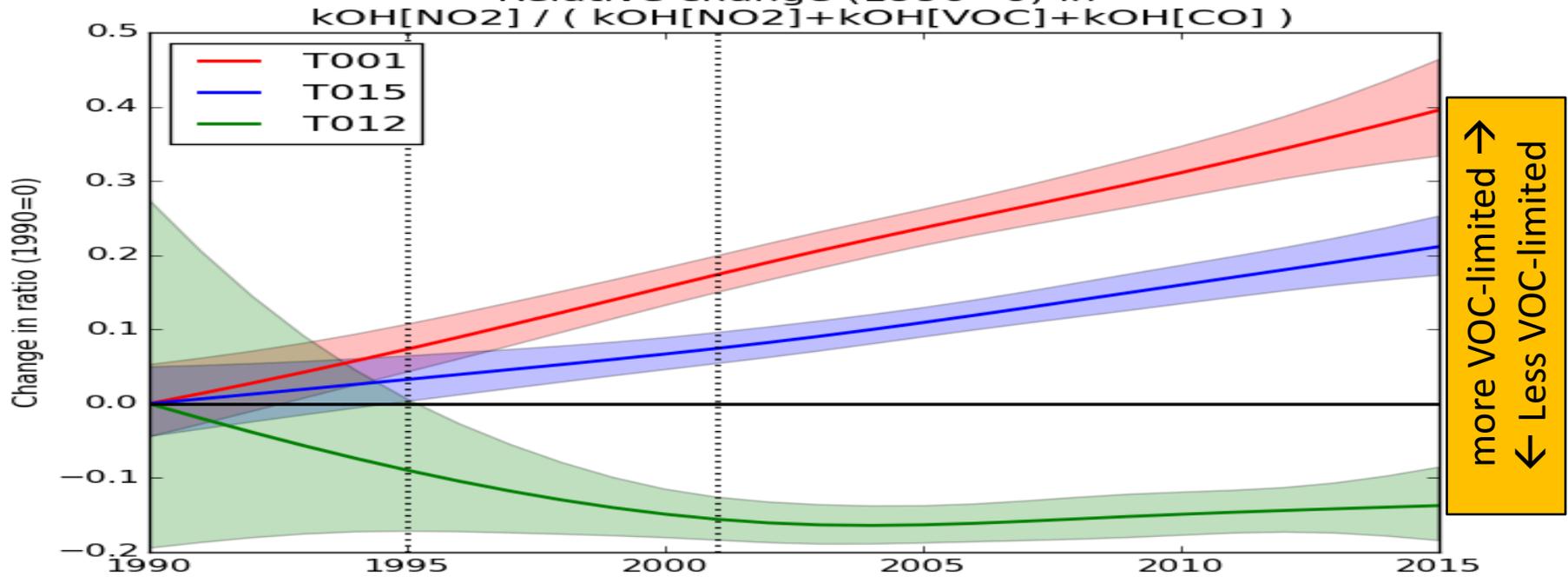
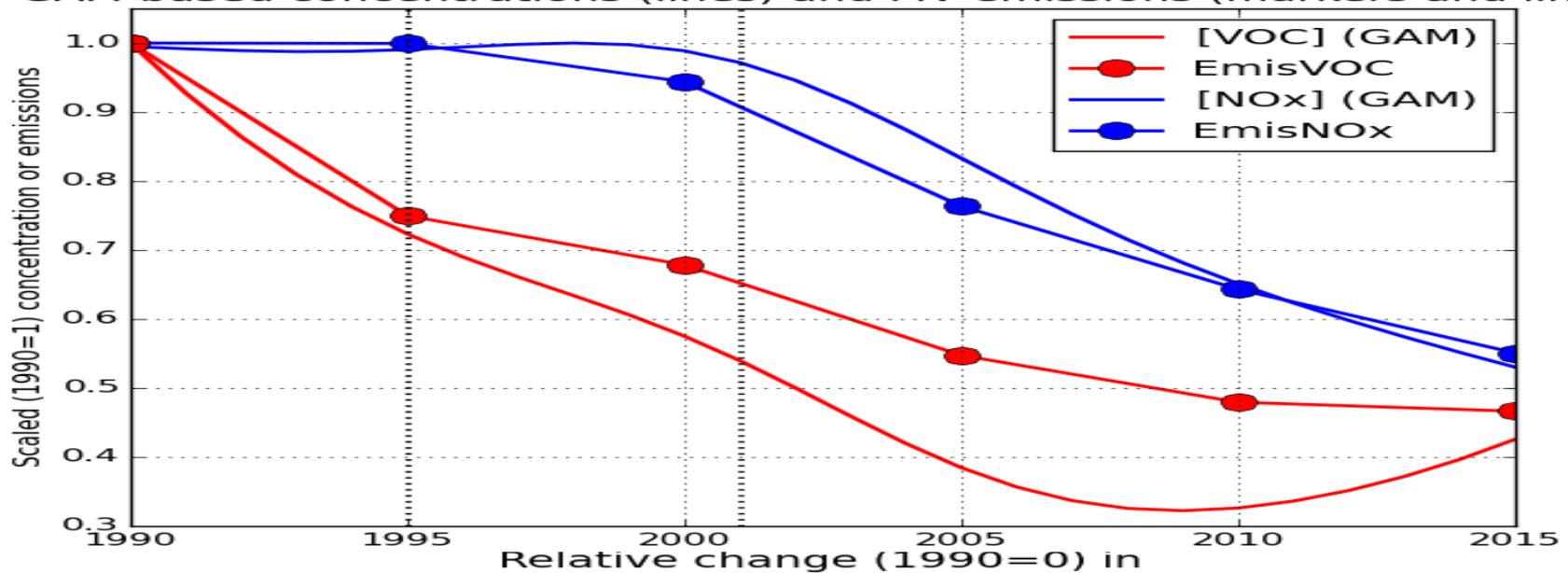
$$H_1 : [O_3] \sim s(Temp, RH) \times s(ETot, DoY) \times s(Ucpt, Vcpt) \times s(WeekDay) \times s(Dist) \times s(Year) + \epsilon$$

Model	df	BIC	AIC
H0	17	-1047	-1125
H1	15	-1060	-1128

It appears so!
Both BIC and AIC scores **do not**
support the more complex H₀
model (with Dist+Year interaction
term).

$$\text{Ratio of } \frac{k_{\text{OH}}[\text{NO}_2]}{(k_{\text{OH}}[\text{NO}_2] + k_{\text{OH}}[\text{CO}] + k_{\text{OH}}[\text{VOC}])}$$

Comparison of average annual VOC and NOx
 GAM-based concentrations (lines) and MV emissions (markers and lines)



Conclusions -- background

- Trends look to be ~ 0.2 ppb/year trend, some suggestion that not uniform
- Not much inter-station variability

Conclusions – fair weather

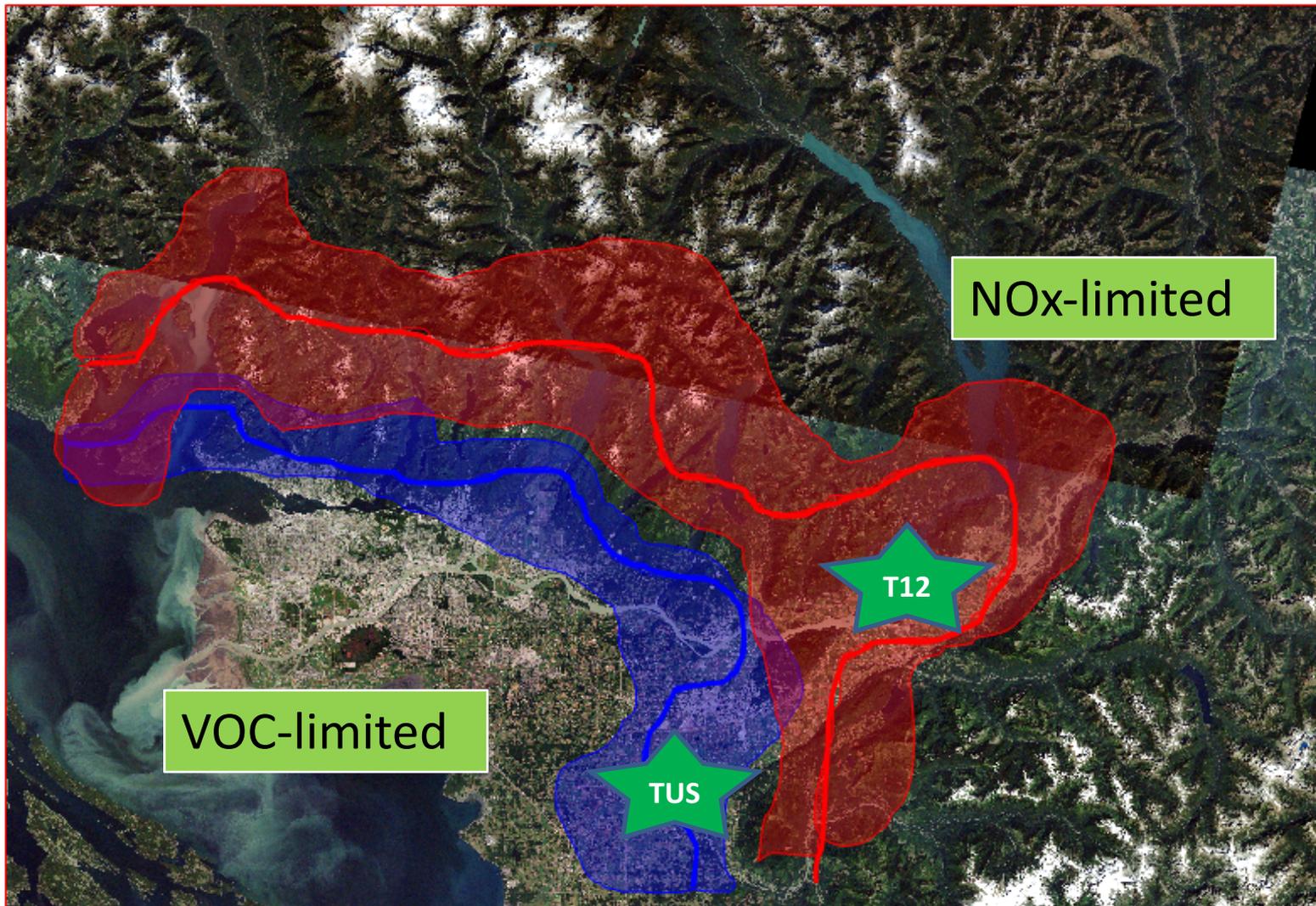
- Trends **not** uniform/linear
- Trends best understood after removing background
- A lot of inter-station variability

Conclusions -- CAAQS

- Results similar to FAIR WEATHER but with bigger trends. Why?
- No inter-station variability. Why?

Future Work

- Are these results consistent with the earlier modeling work?



Hand drawn VOC-to-NO_x transition regions based on CMAQ model output
Coloured regions give estimated extent of variability from varying meteorological conditions
Red colours for CMAQ output using 1985 emissions
Blue colours for CMAQ output using 2005 emissions

Questions?

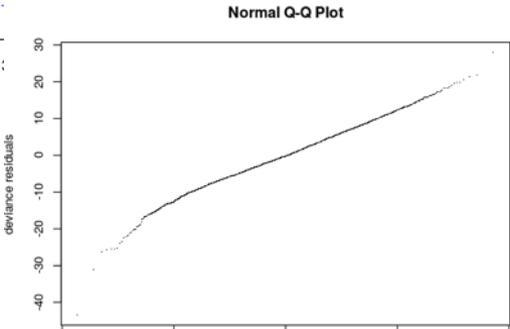
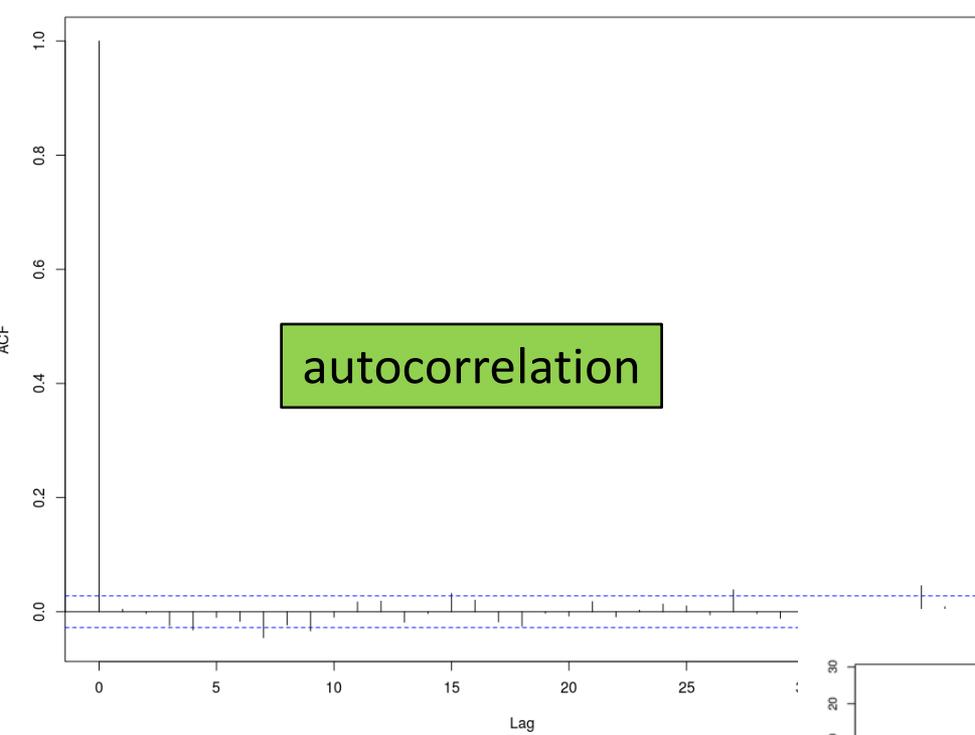
Thanks !

Previous LFV Trend analysis

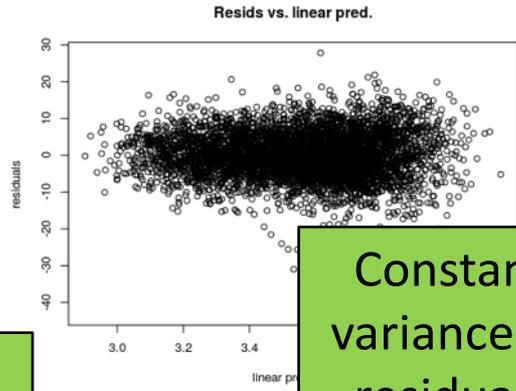
Reference	Timeframe	Comments
Joe et al. (1996)	1978-1990	1-hr ozone; isotonic regression; 2 stations
Pryor (1998)	1984-1991	1-hr ozone; trends in declimatized ozone; 8 stations
Vingarzan and Taylor (2003)	1985-2000	1-hr ozone; GLS regression; 5 stations
Chan (2009)	1997-2006	8-hr ozone; GLMM; 4 stations (as part of larger analysis)
This study (2017)	1990-2014	8-hr ozone; GAM; 9 stations

Summary of GAM fit to ozone candidate days

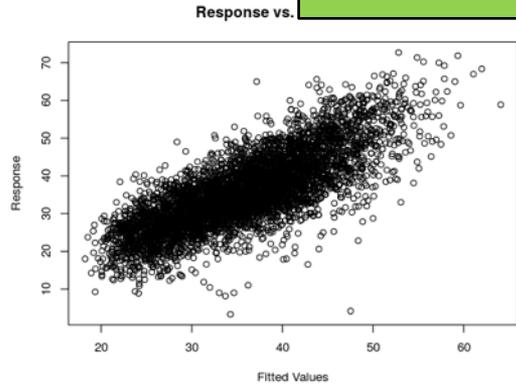
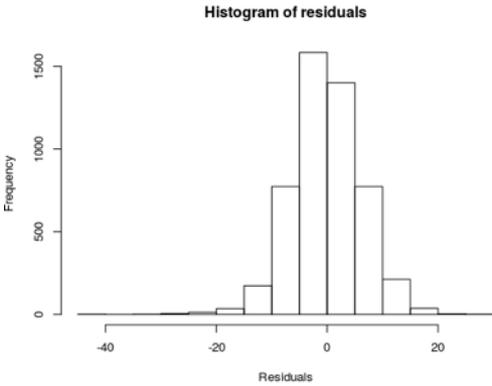
autocorrelation



Normality of residuals



Constant variance of residuals



GAM Model summary

```
> summary(gammobjLAC$gam)
```

```
Family: gaussian  
Link function: log
```

```
Formula:
```

```
Ozone ~ te(cTemp, cRH) + te(cE_tot, cDoY) + s(cUcpt, cVcpt, by = Meso_f) +  
  s(Weekend, bs = "cc", k = 7) + s(cDist, k = 7, bs = "ts") +  
  s(cYear, k = 10, bs = "ts") + ti(cDist, cYear, k = c(7, 10))
```

```
Parametric coefficients:
```

```
      Estimate Std. Error t value Pr(>|t|)  
(Intercept) 3.528496   0.008138   433.6 <2e-16 ***
```

```
---  
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1
```

```
Approximate significance of smooth terms:
```

	edf	Ref.df	F	p-value	
te(cTemp,cRH)	13.683	13.683	95.785	< 2e-16	***
te(cE_tot,cDoY)	11.420	11.420	20.549	< 2e-16	***
s(cUcpt,cVcpt):Meso_f1	16.097	16.097	15.604	< 2e-16	***
s(cUcpt,cVcpt):Meso_f2	9.573	9.573	19.150	< 2e-16	***
s(cUcpt,cVcpt):Meso_f3	16.824	16.824	16.982	< 2e-16	***
s(cUcpt,cVcpt):Meso_f4	11.591	11.591	31.778	< 2e-16	***
s(Weekend)	3.871	5.000	5.110	9.87e-06	***
s(cDist)	5.636	6.000	90.730	< 2e-16	***
s(cYear)	3.545	9.000	2.662	9.30e-06	***
ti(cDist,cYear)	5.292	5.292	2.522	0.027	*

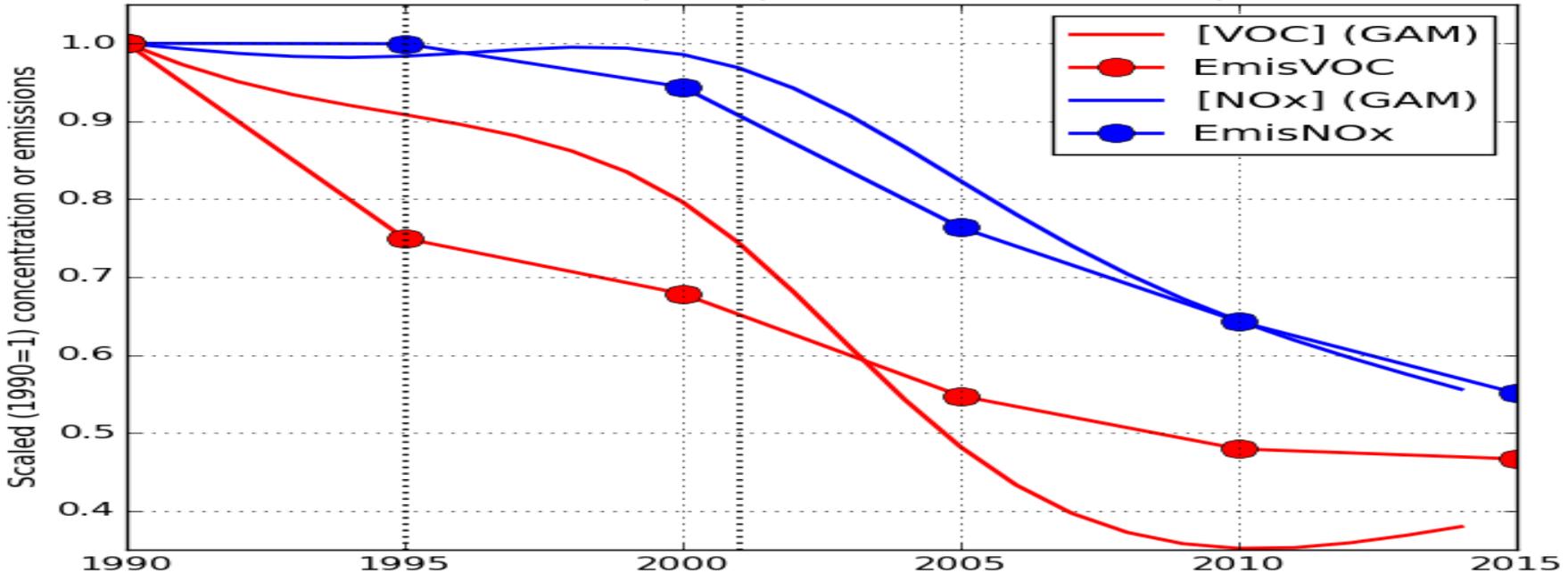
```
---  
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1
```

```
R-sq.(adj) = 0.603
```

```
Scale est. = 40.967    n = 5009
```

Comparison of average annual VOC and NOx

GAM-based concentrations (lines) and MV emissions (markers and lines)



Relative change (1990=0) in ozone CANDIDATE over time

