Radiocarbon as a tracer for the source and fate of fossil fuel CO_2 emissions



Jocelyn Turnbull

Rafter Radiocarbon Lab, GNS Science, New Zealand
and University of Colorado, Boulder, USA

Nikita Turton, Lucas Domingues, and the Rafter Radiocarbon Team
Ken Davis, Natasha Miles, Scott Richardson, Anna Karion, Isaac Vimont

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Outline

- Introduction
- Tracing fossil fuel CO_2 (CO_2ff) with radiocarbon
  - Indianapolis example
- The Great Greenhouse Gas Grass-off
  - Covid-19 related CO_2ff emission reductions in New Zealand

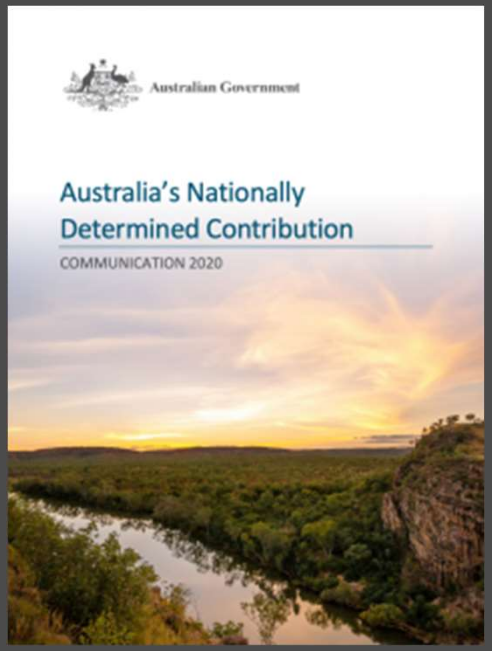
All nations have committed to reducing their greenhouse gas emissions

The Paris Agreement

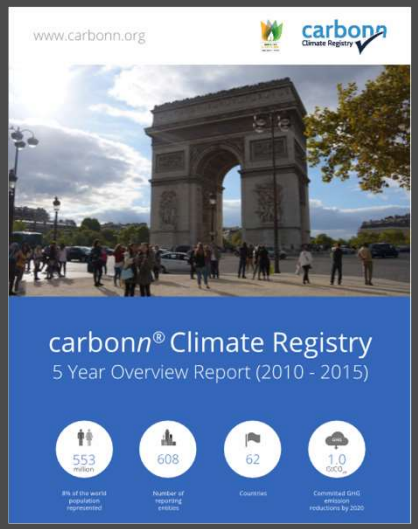
United Nations
Framework Convention on Climate Change

National Mitigation Targets

ZERO CARBON ACT NZ

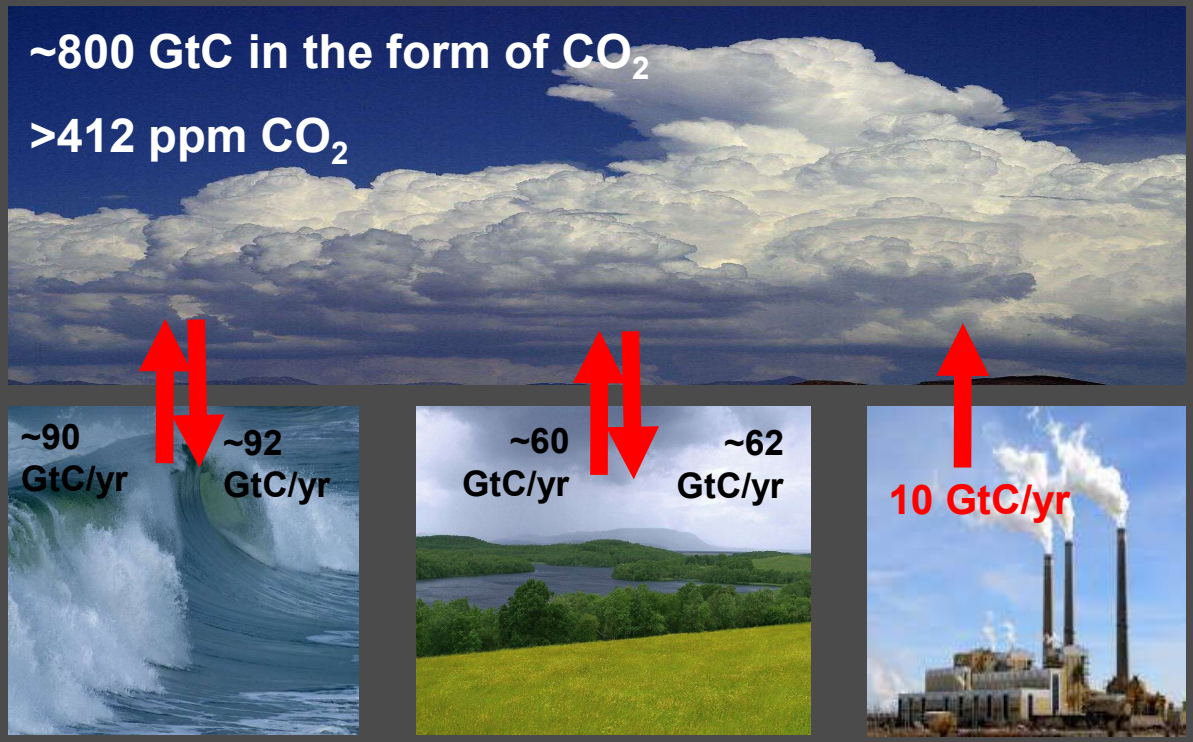


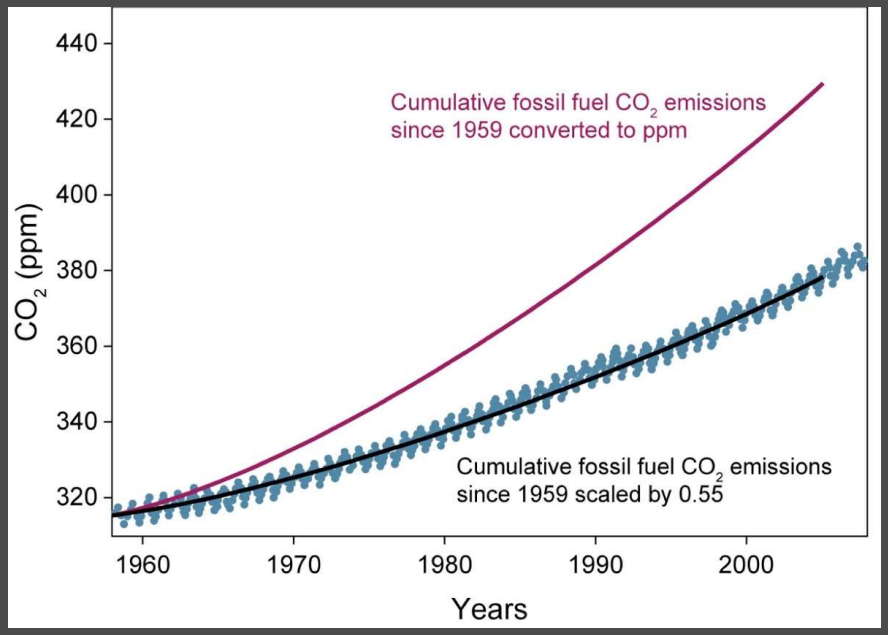
Cities are often leading the way in emission reduction efforts



Policy *decisions* may be at national level
Policy *actions* are largely at the local level
Urban areas produce ~75% of all GHG emissions
Co-benefits of reduced emissions:
Cleaner air, reduced traffic congestion, improved energy security
Emissions information is needed at the city scale

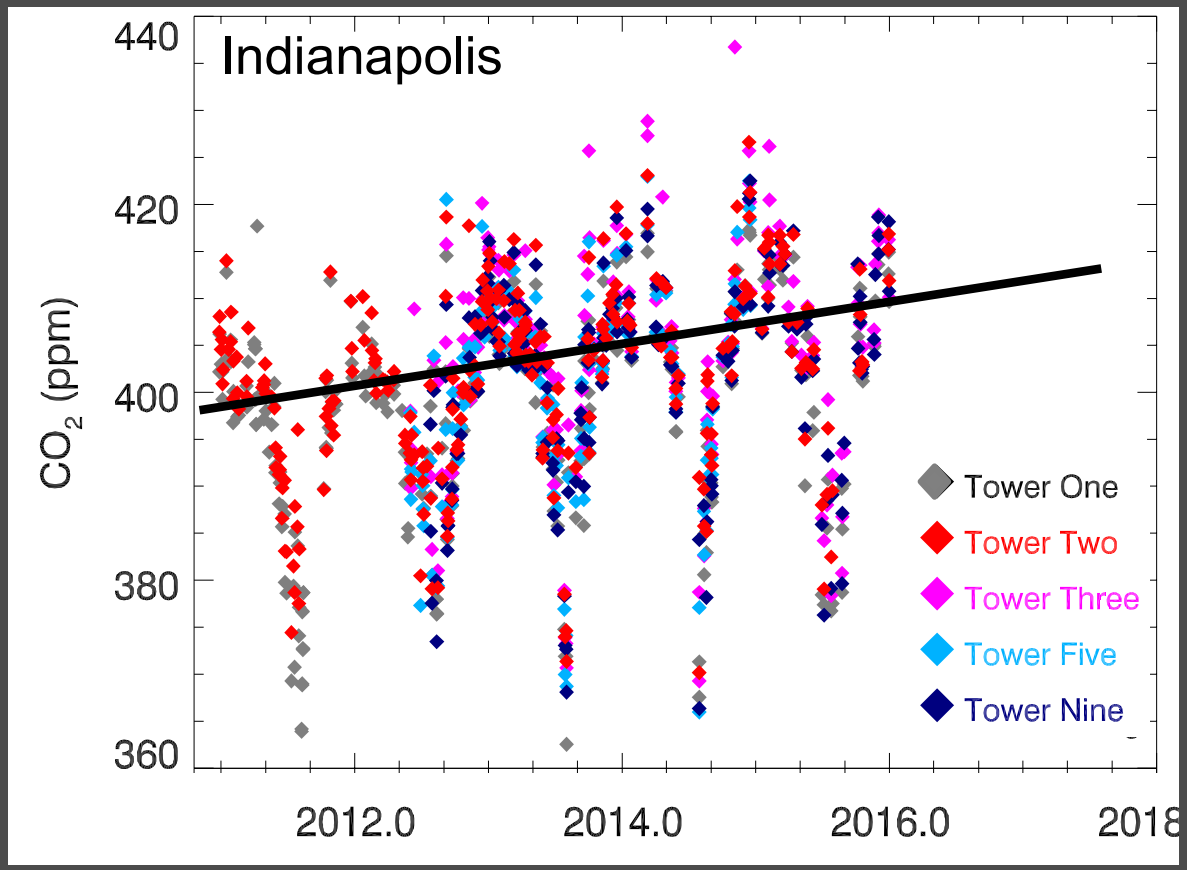
The Carbon Cycle

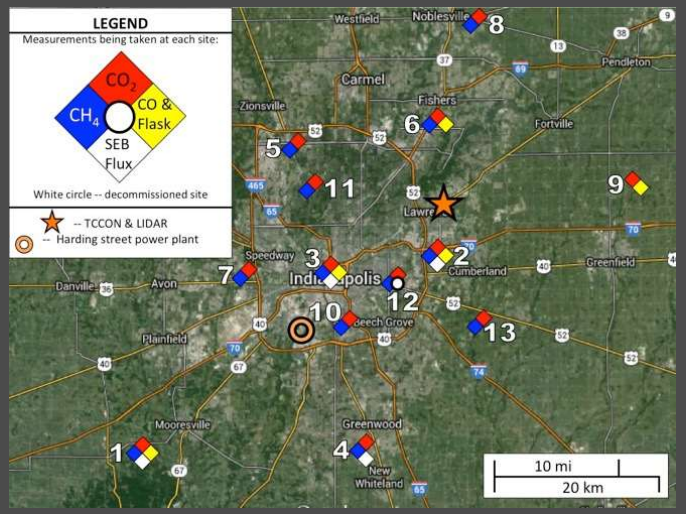




Land and ocean sinks remove about half of all fossil fuel derived CO_2 from the atmosphere

Detecting urban CO_2ff emission rates is challenging with CO_2 measurements alone





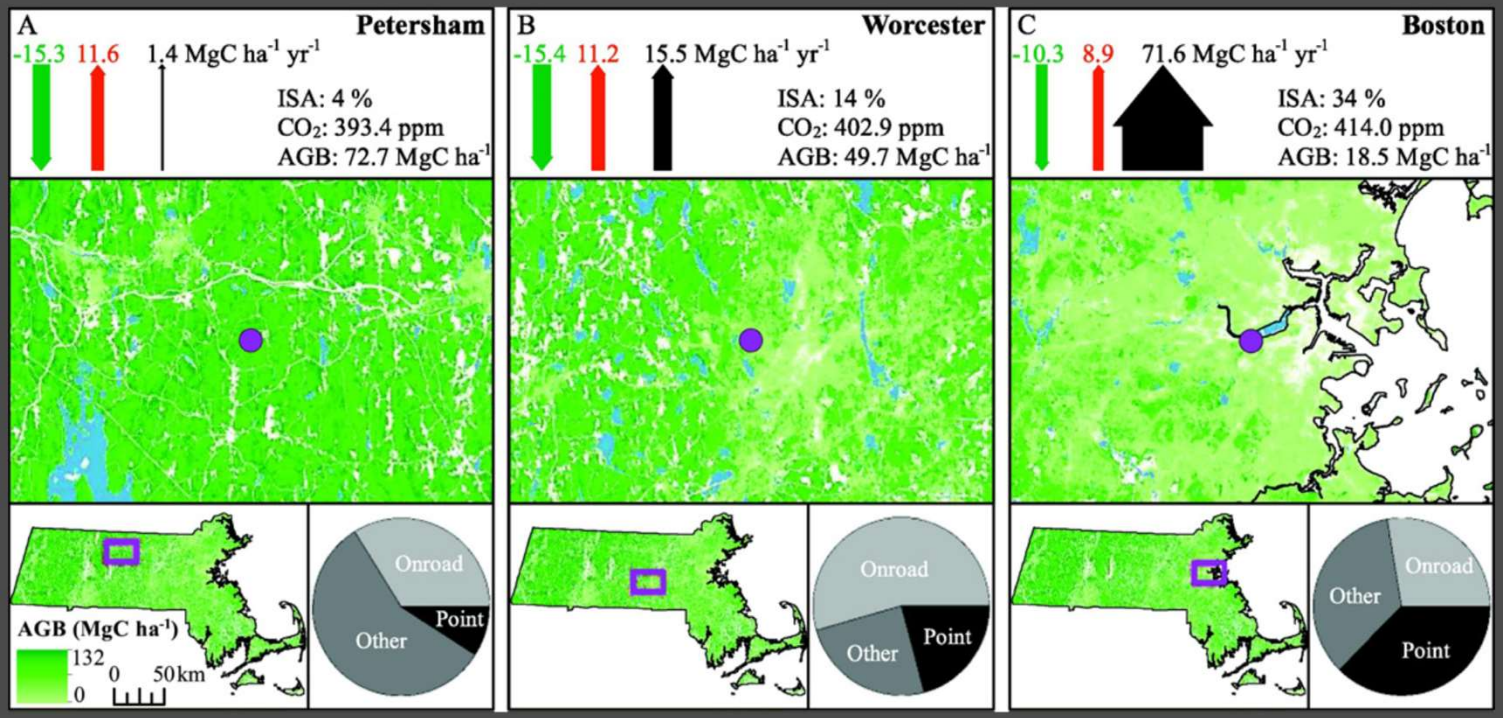
Long-term CO_2 trend due to global fossil CO_2 additions

Seasonal cycle due to photosynthesis and respiration

Local fossil fuel CO_2 signal is difficult to isolate

Turnbull et al., 2015

Urban biogenic (CO_2bio) fluxes

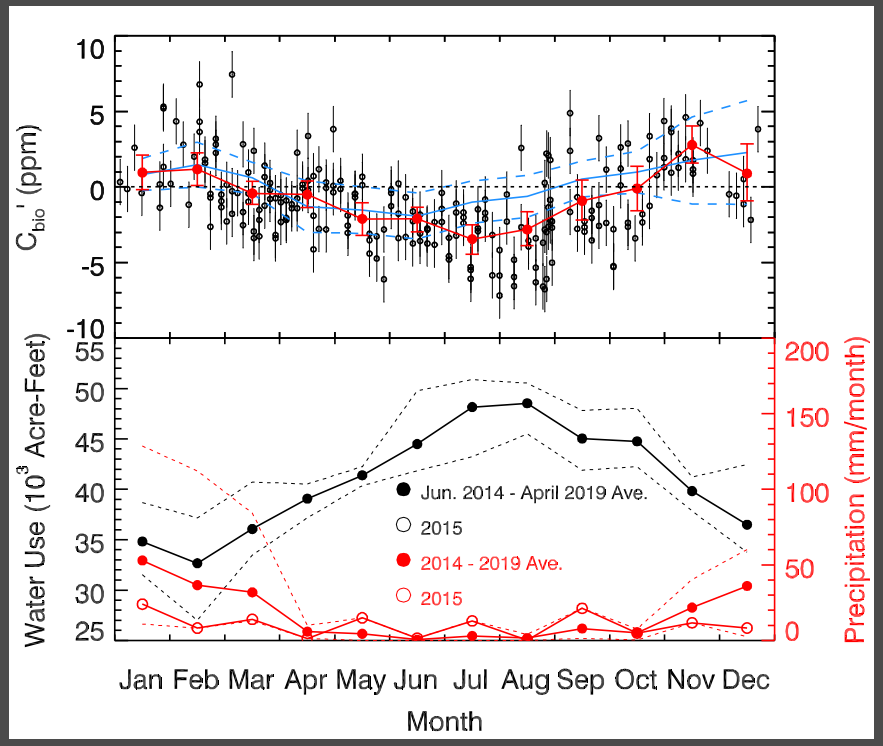


Hardiman et al., 2017

Urban biogenic fluxes are understudied
Can be substantial even in urban core
Variable seasonally and diurnally

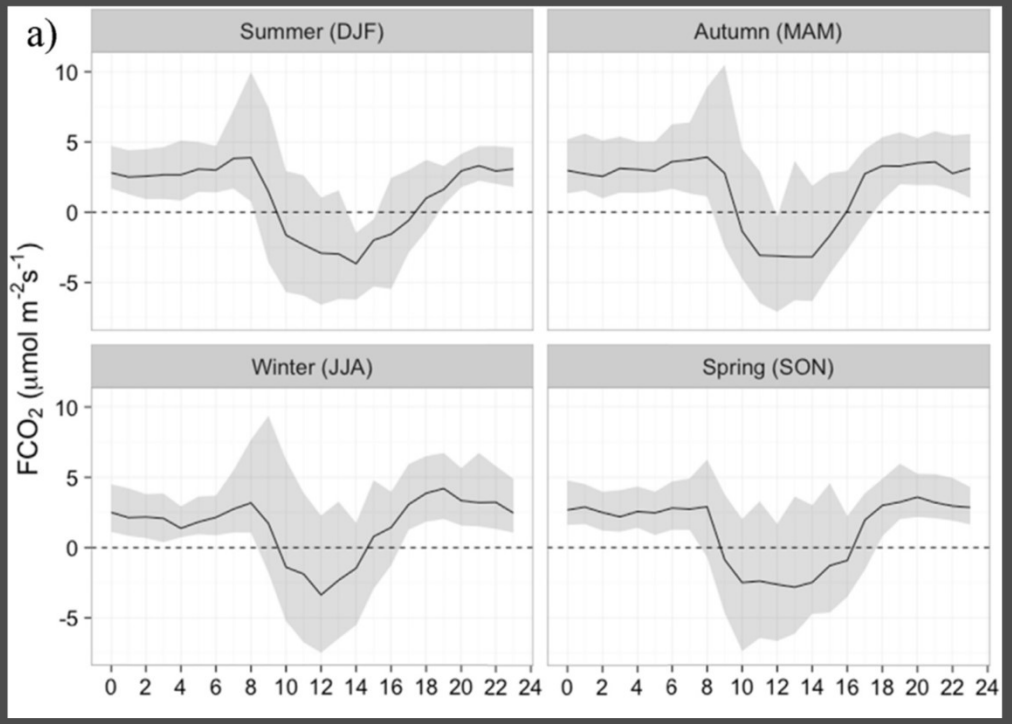
Urban biogenic (CO_2bio) fluxes

Los Angeles



Miller et al., 2020

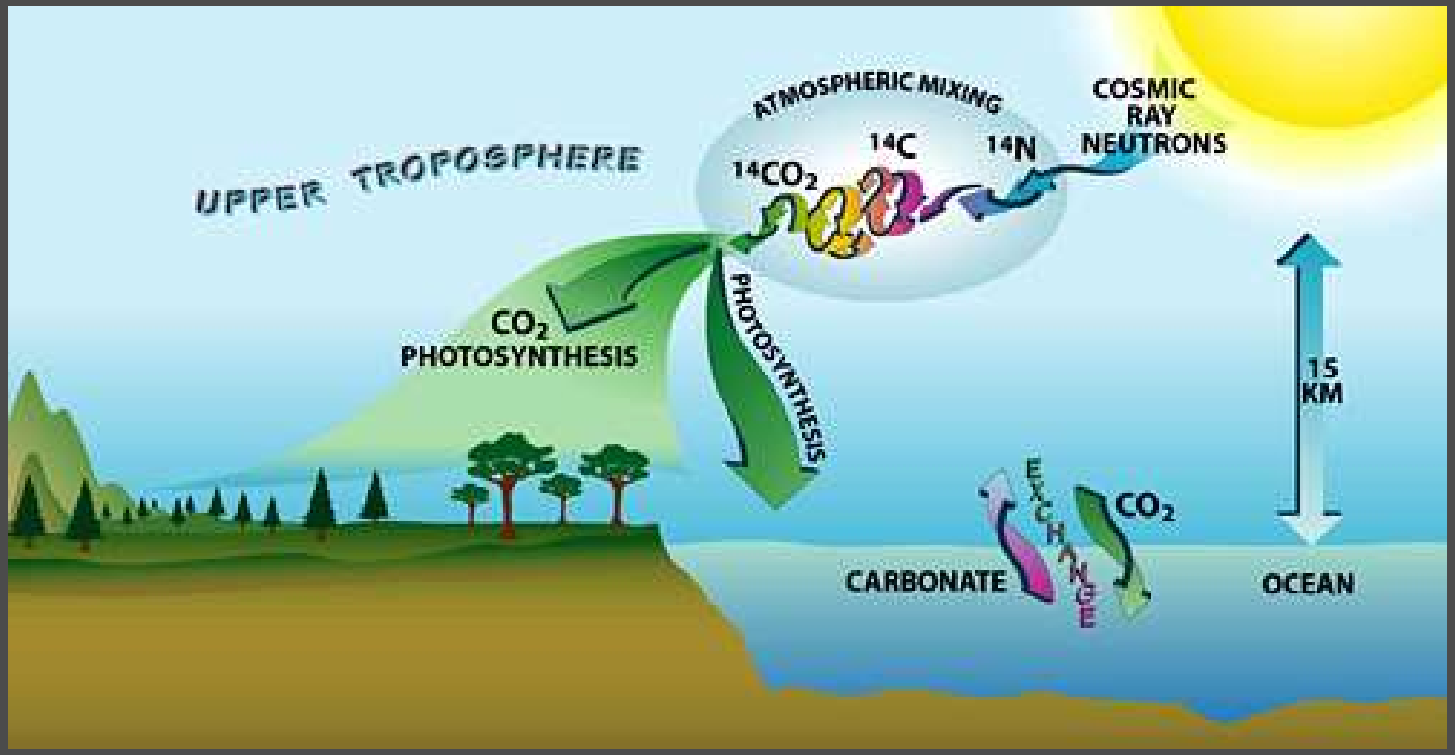
Auckland

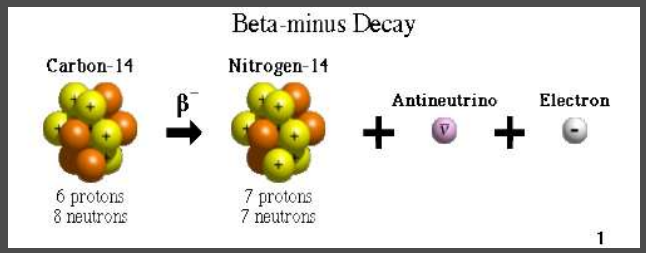


Weissert et al., 2016

CO_2bio contribution varies seasonally and diurnally
Seasonality can be surprising

Radiocarbon (^{14}C) dating





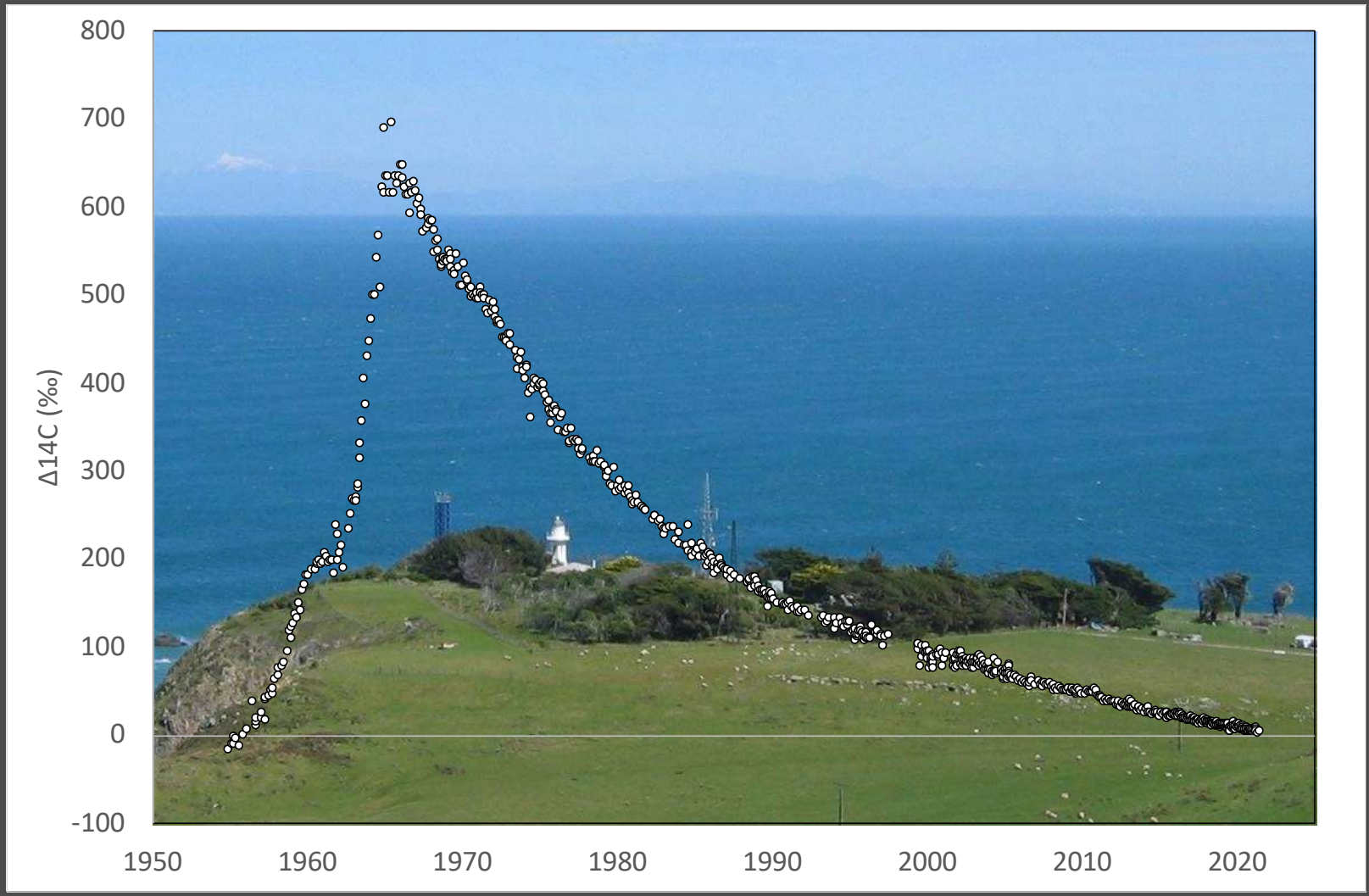
^{14}C is produced naturally in the atmosphere, and moves throughout the carbon cycle

Natural radioactive decay removes ^{14}C from buried/dead objects

Half-life 5,730 years

Fossil fuels are entirely devoid of ^{14}C

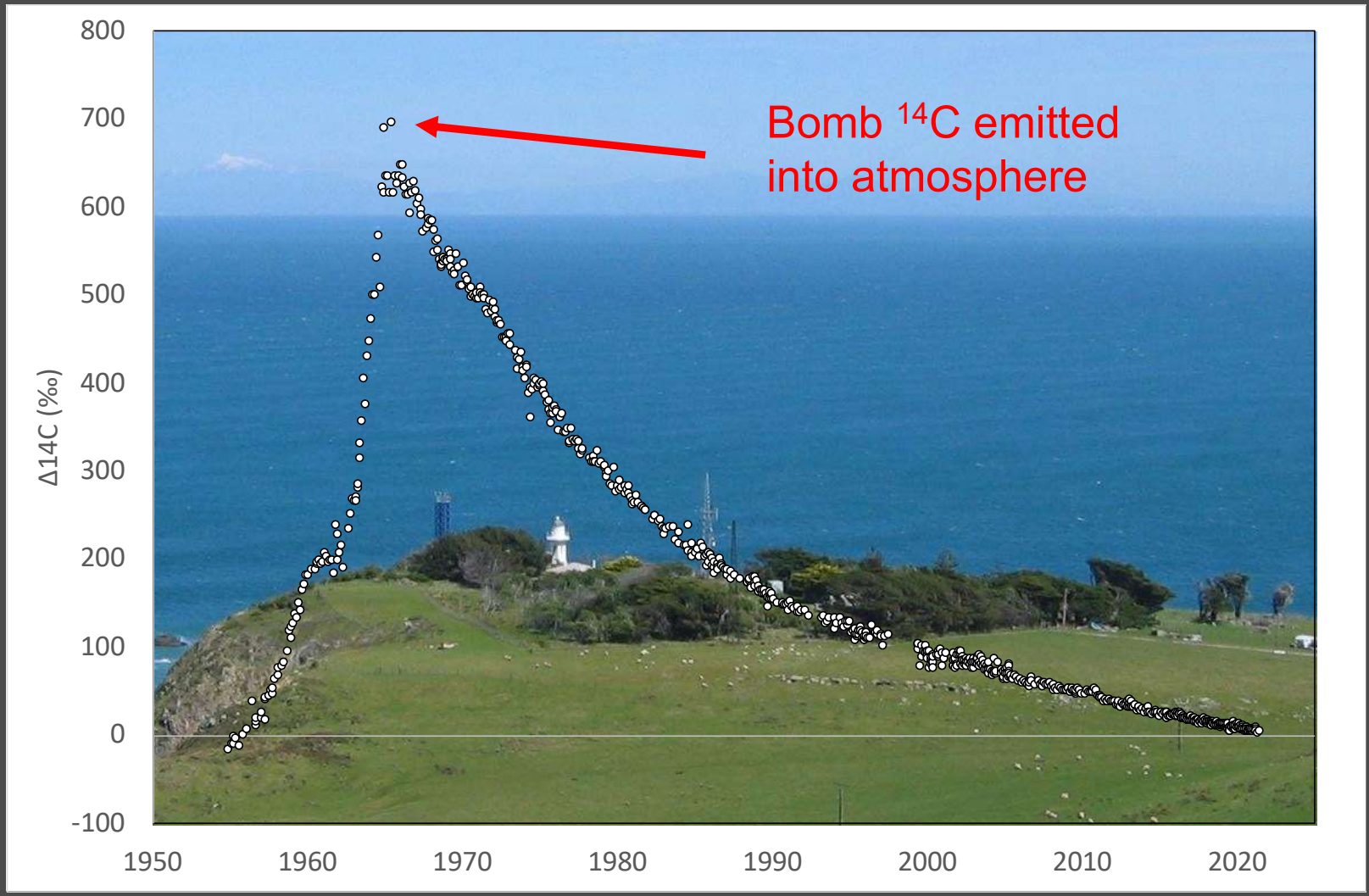
Modern atmospheric radiocarbon history



Wellington $^{14}CO_2$ record 1954 – present

Turnbull et al., 2017

Modern atmospheric radiocarbon history



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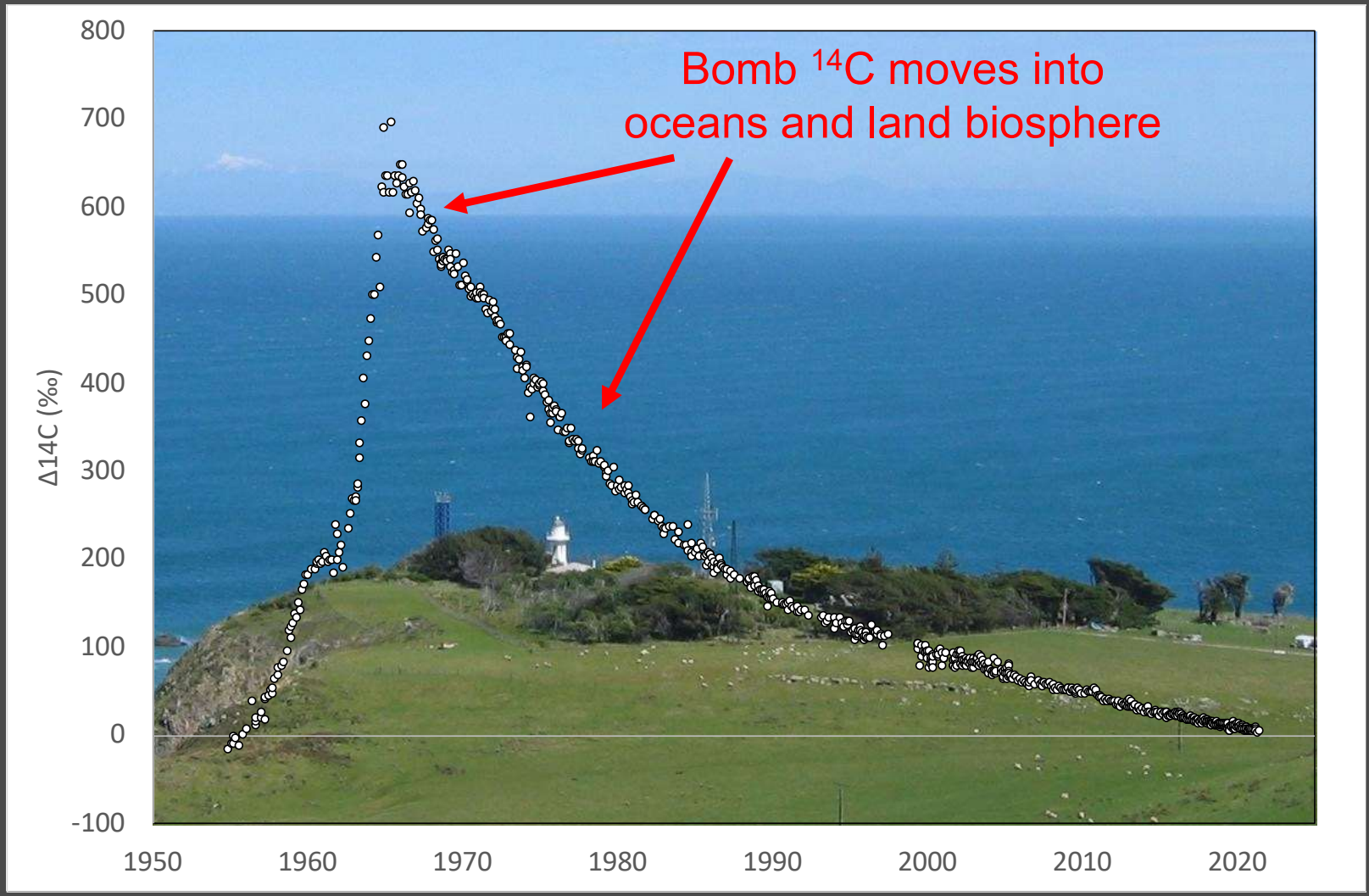

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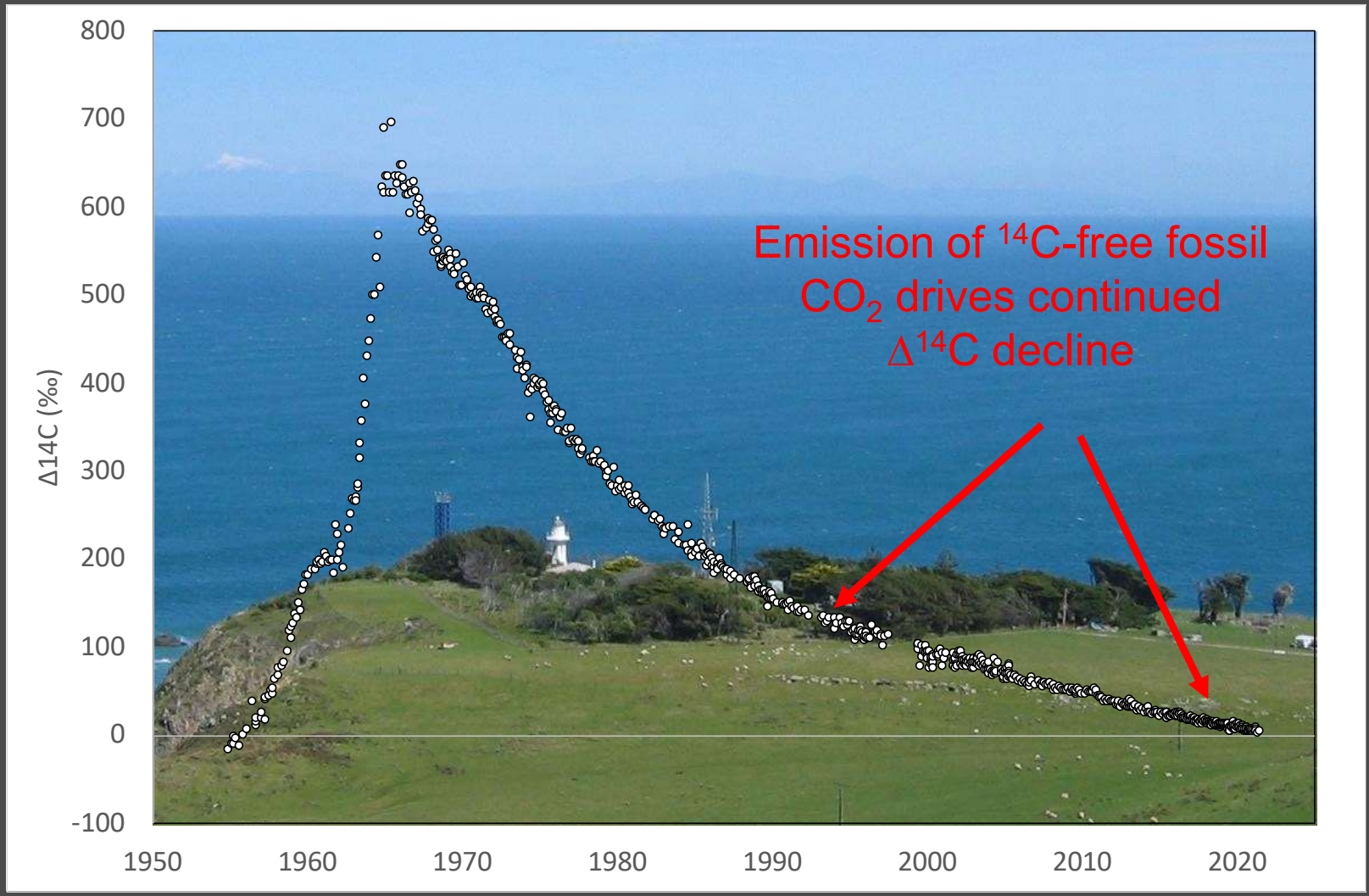
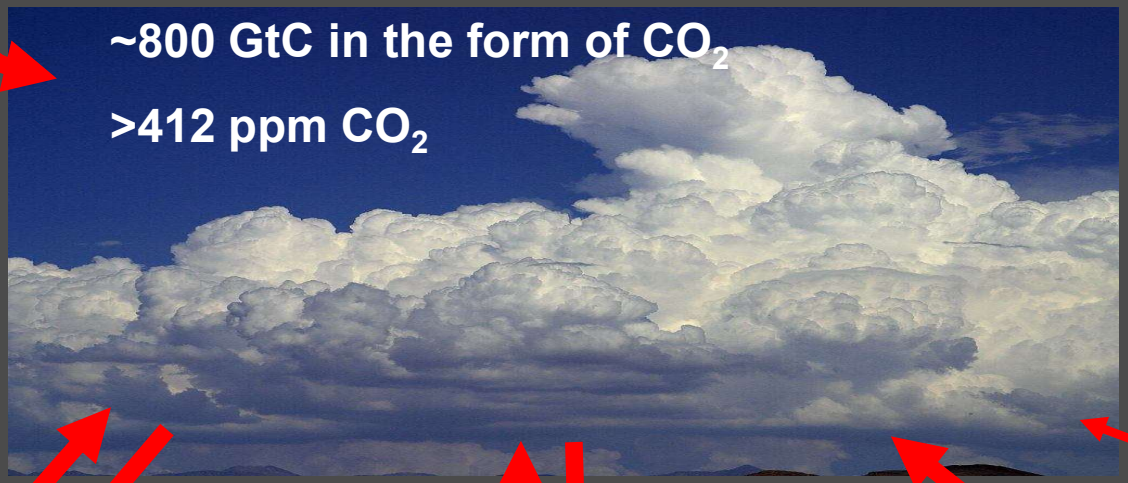


Wellington $^{14}CO_2$ record 1954 – present

Turnbull et al., 2017

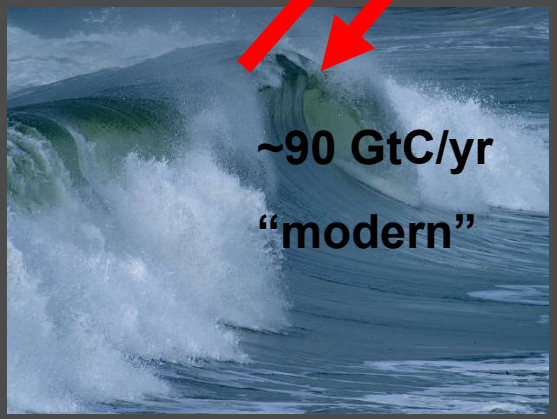
The Radiocarbon Cycle

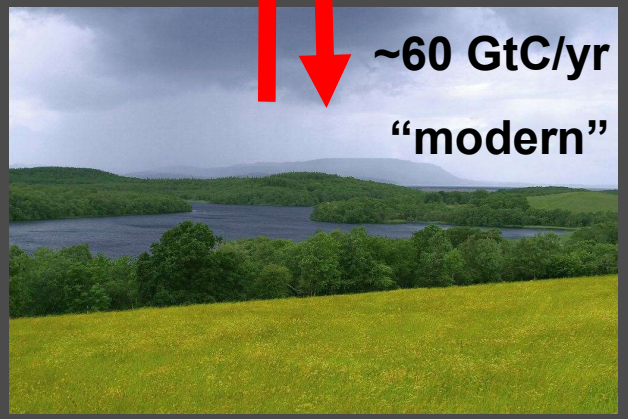
Cosmogenic production Of ^{14}C



$^{14}CO_2$ separates fossil fuel CO_2 from natural CO_2

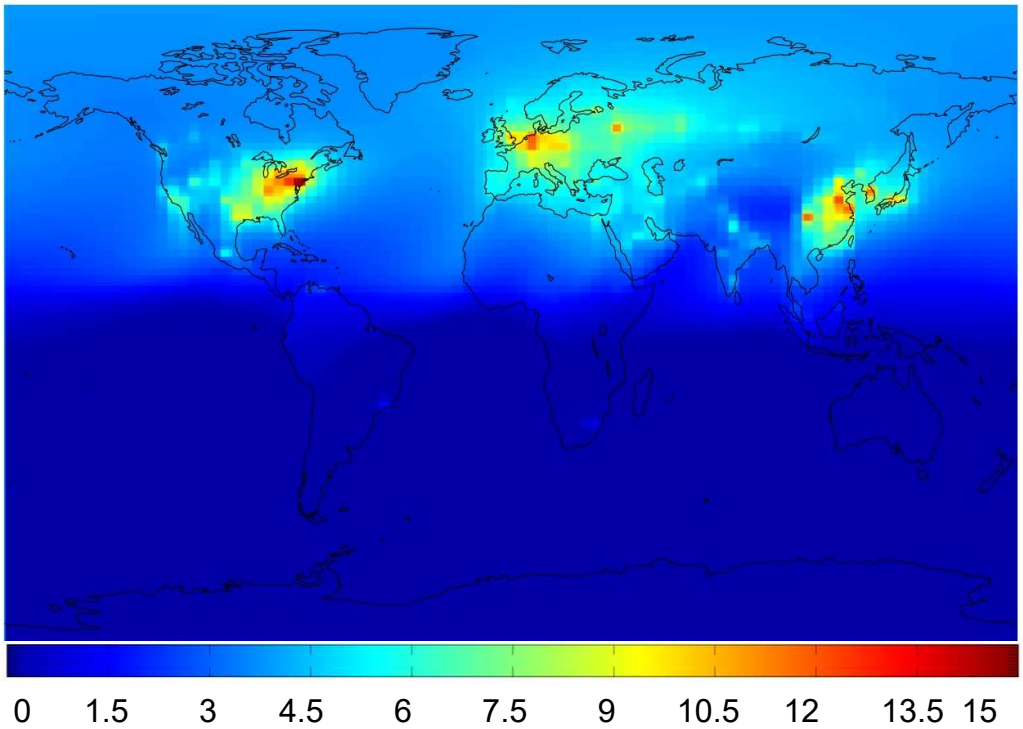
-2.6‰ change in $\Delta^{14}C$ for each 1 ppm fossil fuel CO_2 added

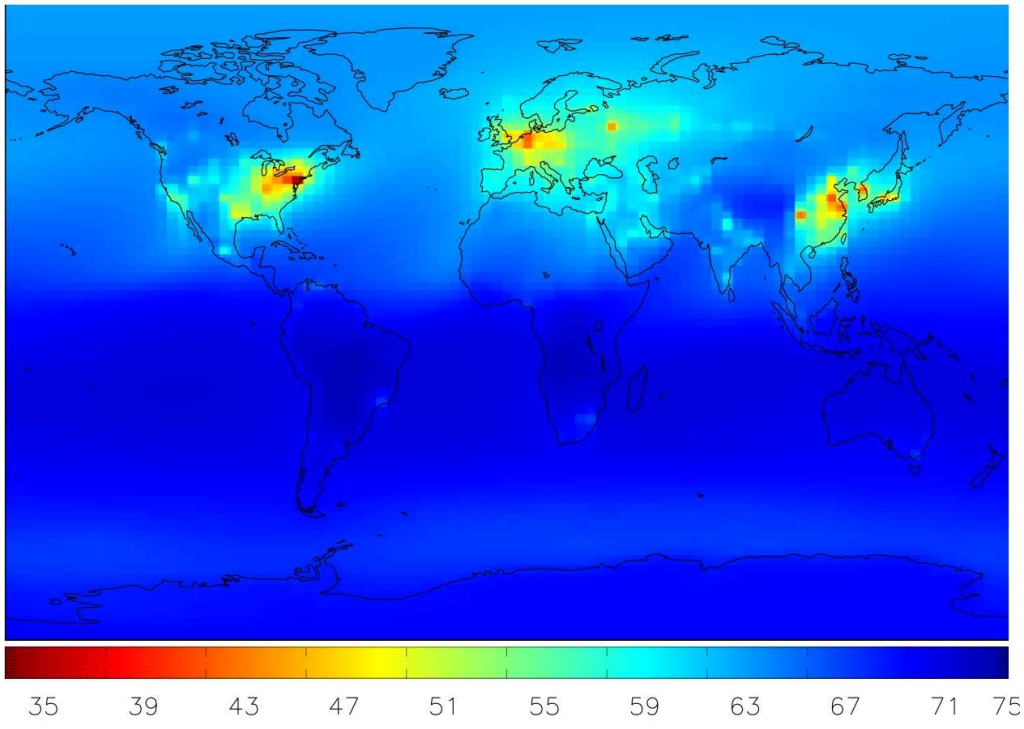




$$CO_2ff = \frac{CO_{2obs}(\Delta_{obs} - \Delta_{bg})}{\Delta_{ff} - \Delta_{bg}} - \frac{CO_{2r}(\Delta_r - \Delta_{bg})}{\Delta_{ff} - \Delta_{bg}}$$

Modern global surface distribution of radiocarbon



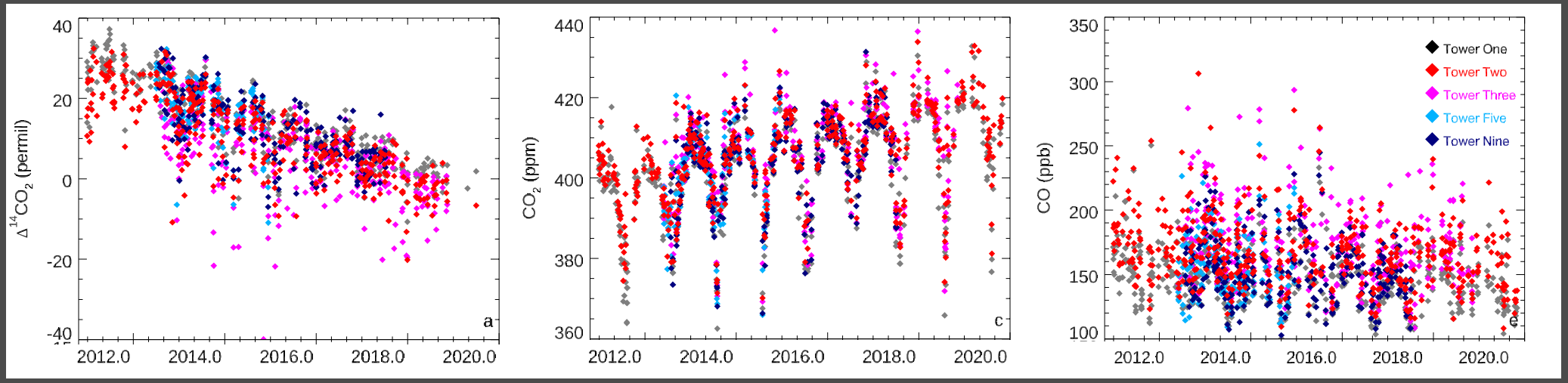


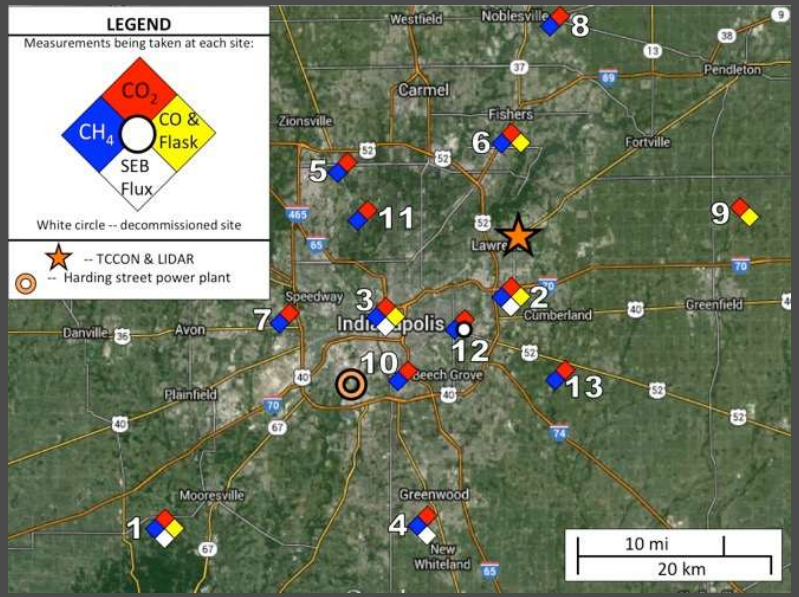
Fossil fuel CO_2 emission pattern very strongly reflected in $\Delta^{14}CO_2$

$$\mathbf{CO_2ff} = \frac{\mathbf{CO_{2obs}\,(\Delta_{obs} - \Delta_{bg})}}{\mathbf{\Delta_{ff} - \Delta_{bg}}} - \frac{CO_{2r}\,(\Delta_r - \Delta_{bg})}{\Delta_{ff} - \Delta_{bg}}$$

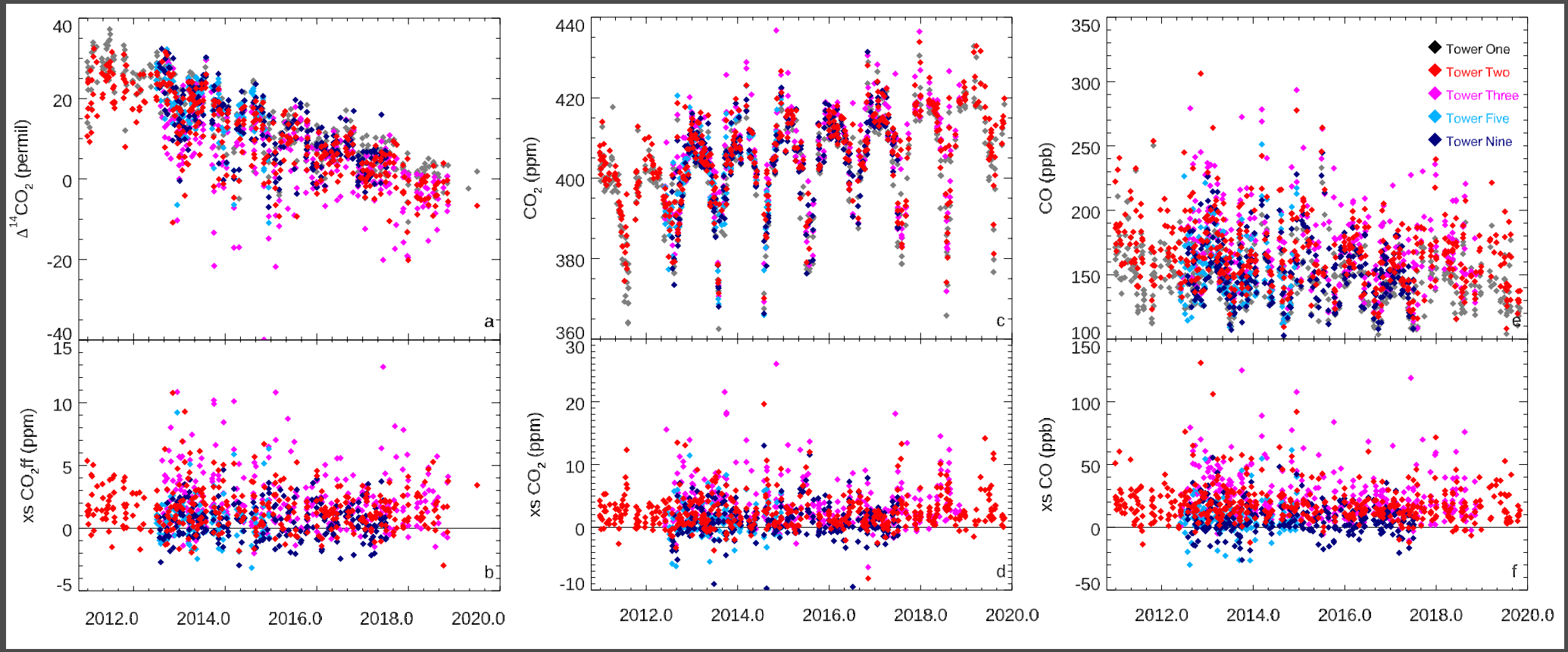
Turnbull et al., 2009
LMDZ model 2002-2007 surface $\Delta^{14}CO_2$

Evaluating urban CO_2ff emissions Indianapolis Flux Project (INFLUX)





INFLUX : Flask-based estimates of total CO_2, CO and CO_2ff



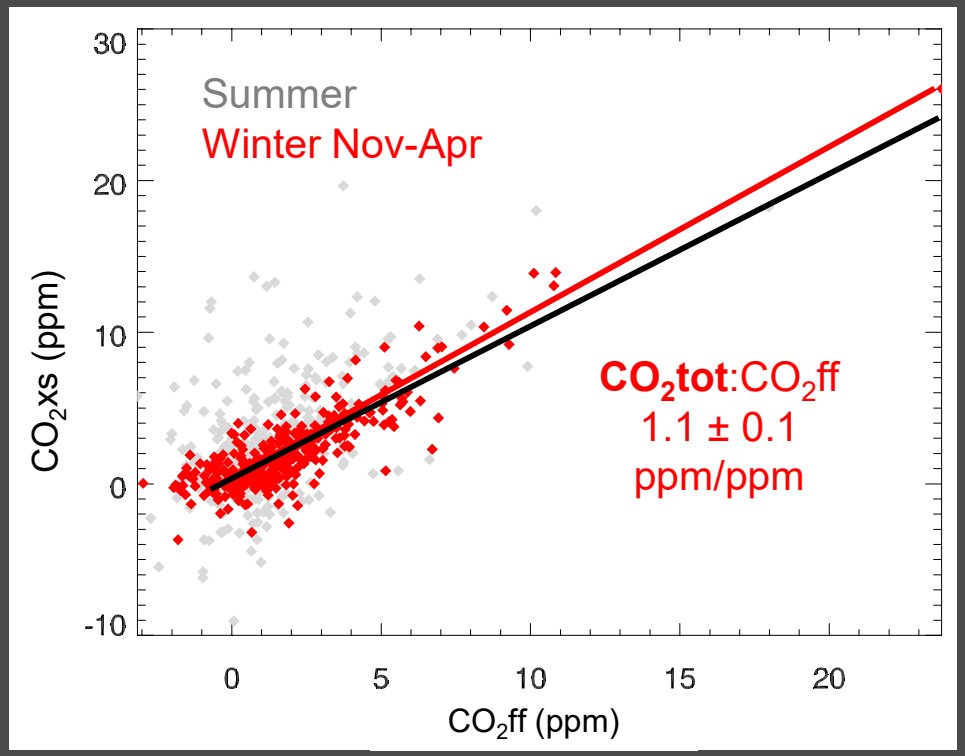
$$CO_2ff = \frac{CO_{2obs}(\Delta_{obs} - \Delta_{bg})}{(\Delta_{ff} - \Delta_{bg})} - ß$$

$CO_2xs = CO_2obs - CO_2bg$

$CO_{xs} = COobs - CObg$

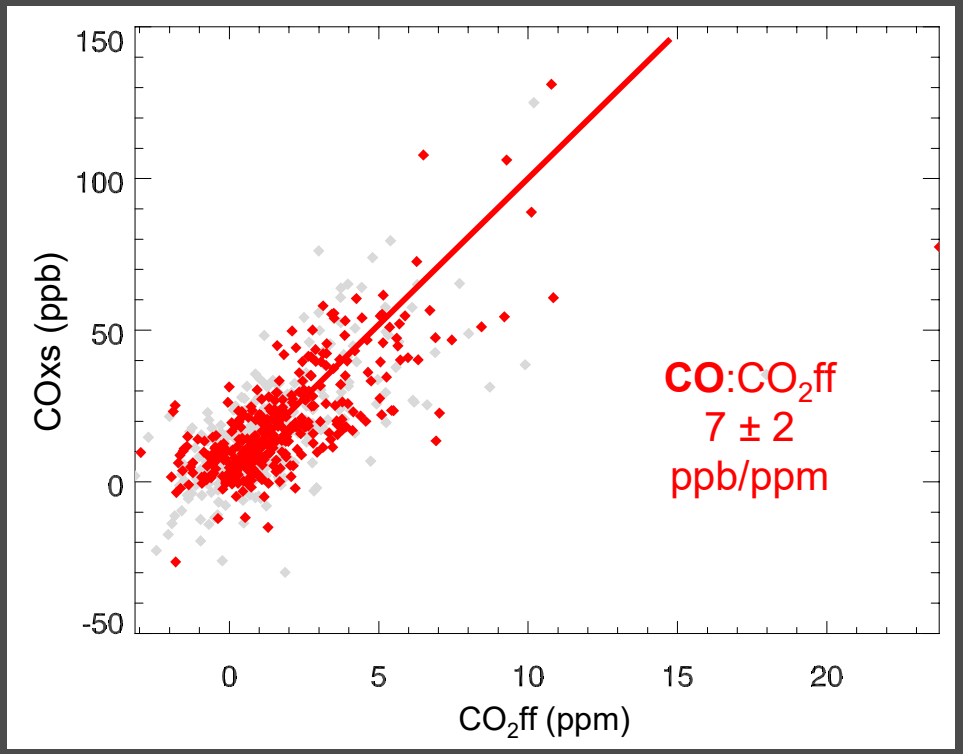
Determine enhancements relative to upwind background Tower One
Consistent enhancements in anthropogenic species at downwind towers

Flask-based emission ratios



Determine how much of the CO_2 comes from fossil fuels and how much from other sources (plant photosynthesis/respiration, human/pet respiration, biomass burning)

~10% contribution of non-CO_2ff to CO_2 in winter

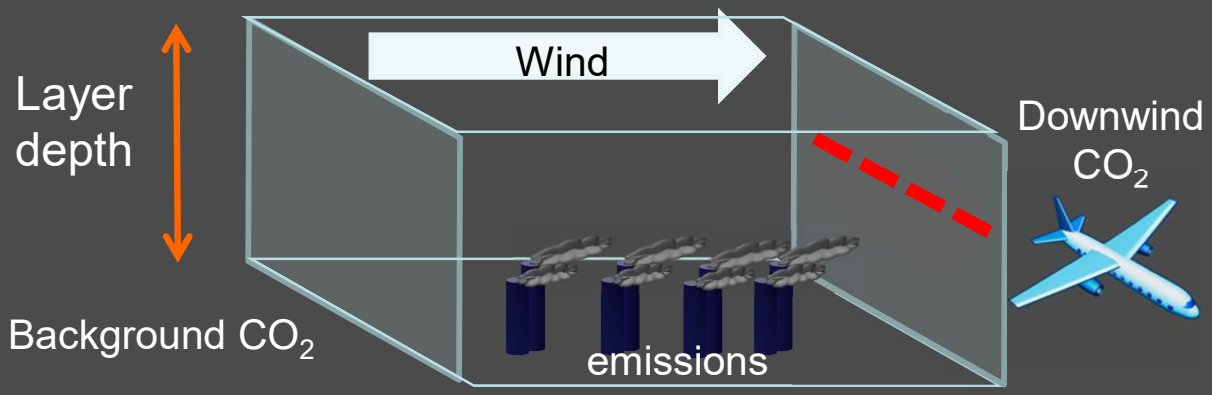


CO co-emitted with CO_2ff at variable rate depending on combustion conditions - derive ratio empirically from observations

Can then use high resolution CO observations to determine CO_2ff

Turnbull et al., 2015

Urban mass balance from aircraft measurements

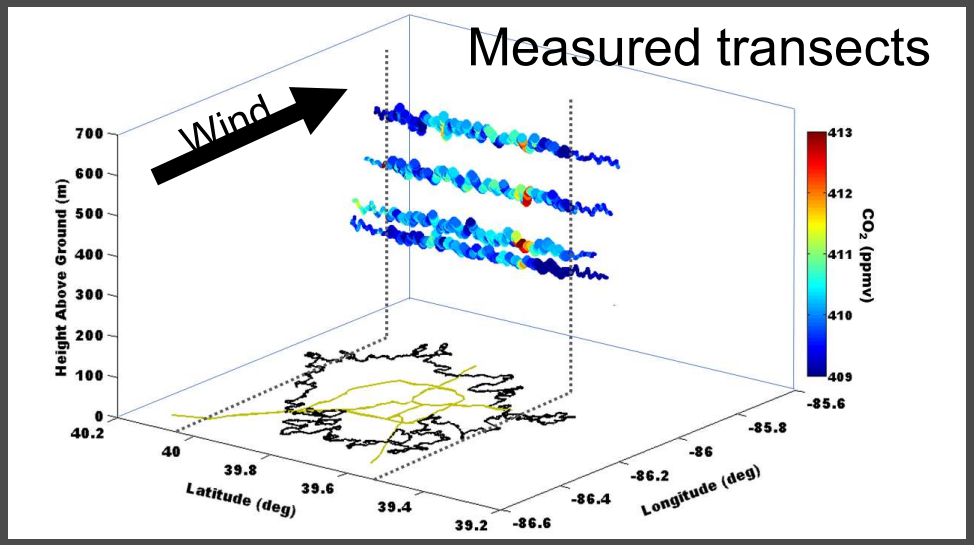


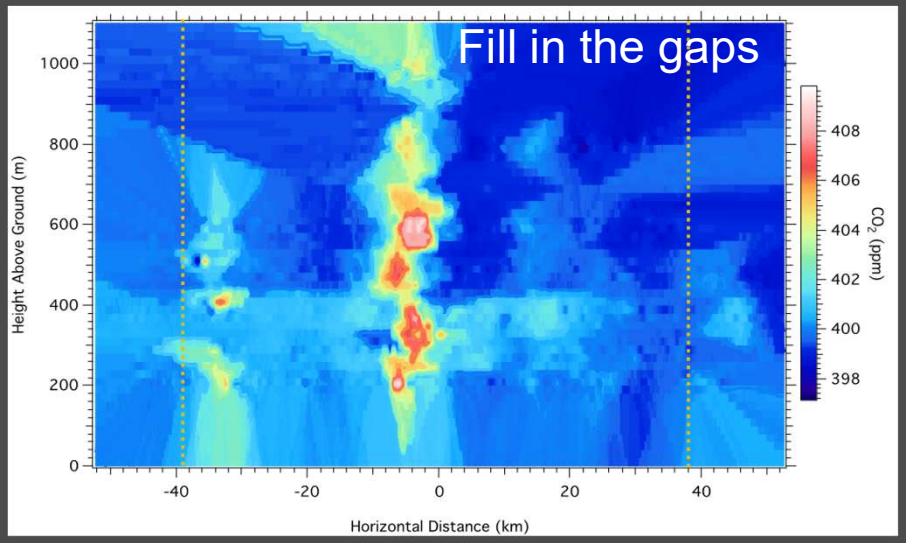
Molar enhancement in air layer

Flux

$$\dot{n}_{CO_2} = V \cos\theta \int_{-b}^{+b} \Delta X_{CO_2} \left(\int_{z_{gnd}}^{z_{PBL}} n_{air} dz \right) dx$$

Perpendicular wind speed

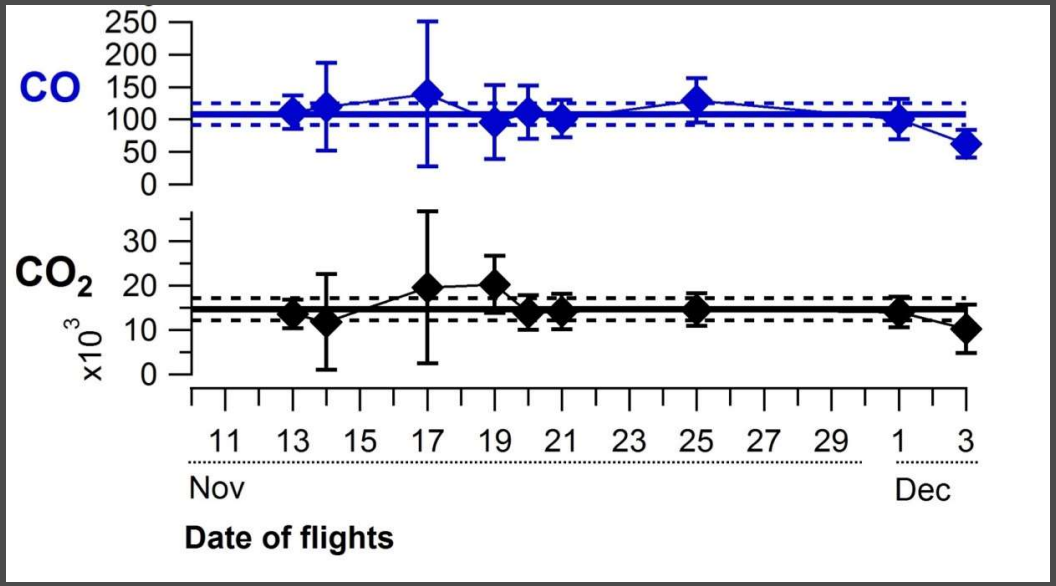




Many cities can be measured using a single aircraft/instrument
Whole city flux determined – not spatially resolved

Heimburger et al., 2017

Urban mass balance from aircraft measurements

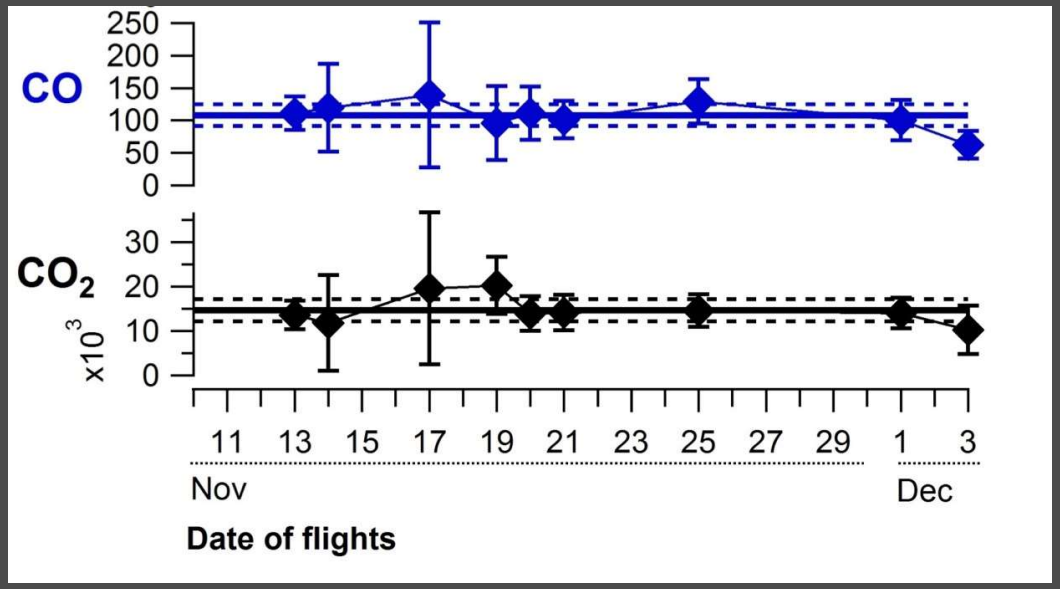


	Emission rate (mol/s)
CO_2	14,600 ± 17%
CO	108 ± 16%

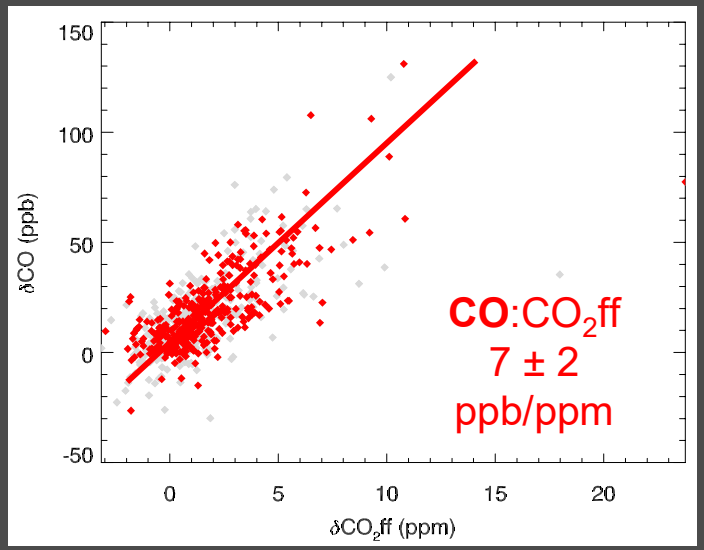
Determine CO_2 and CO emission rates from mass balance

Heimburger et al., 2017

Urban mass balance from aircraft measurements



	Emission rate (mol/s)
CO_2	14,600 ± 17%
CO	108 ± 16%
CO_2ff from CO/^{14}C	13,400 ± 16%



Determine CO_2 and CO emission rates from mass balance

Use ^{14}C-based CO:CO_2ff to calculate Indianapolis' CO_2ff emission rate

Heimburger et al., 2017
Turnbull et al., 2019

The Great Greenhouse Gas Grassoff

Bored kids collecting grass clippings during lockdown help reveal dramatic cuts to carbon emissions

Olivia Wannan · 05:00, Aug 20 2020

stuff.co.nz

Citizen science campaign to collect grass samples during lockdown

Plants use photosynthesis to absorb CO_2 from the air and use it to grow

The radiocarbon content of that CO_2 is faithfully recorded in the leaves

Can determine the average CO_2ff content the air for the growth period of the leaf

Great Greenhouse Gas Grassoff

441 participants signed up

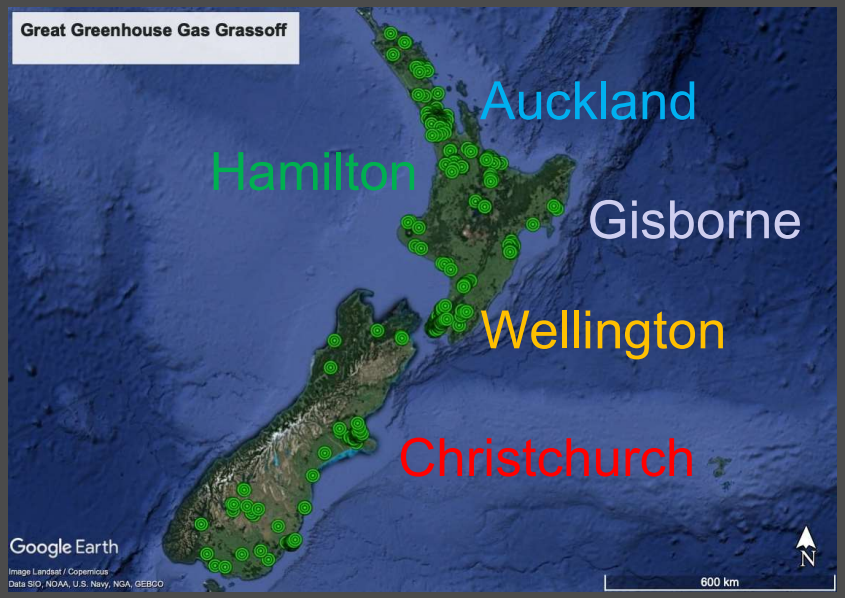
Grass samples received from 110 citizens including lots of children

Selected sites with ~weekly samples through Level 4 to Level 1

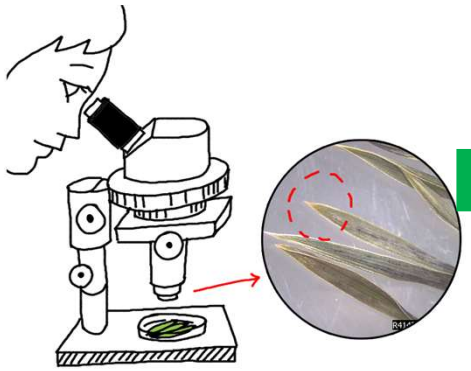
Screening for sample quality

Screening for sites with detectable CO_2ff signals

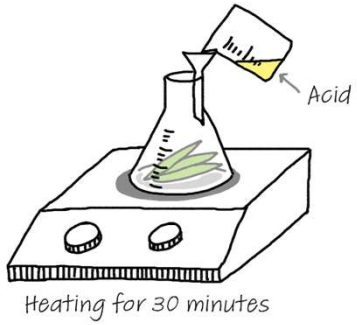
17 sites with usable data in five cities



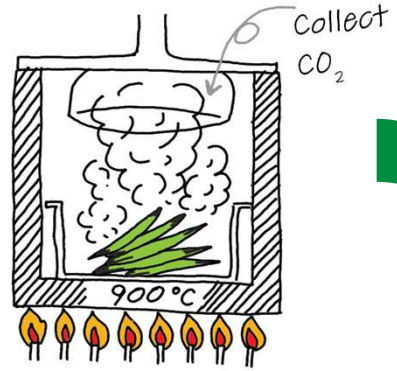
Measurement method



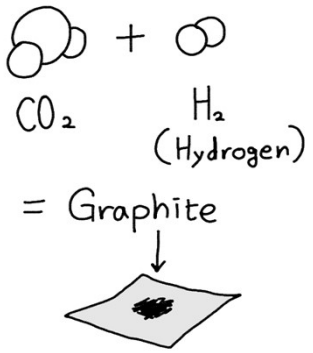
1. Select the grass sample with the most recent growth point.



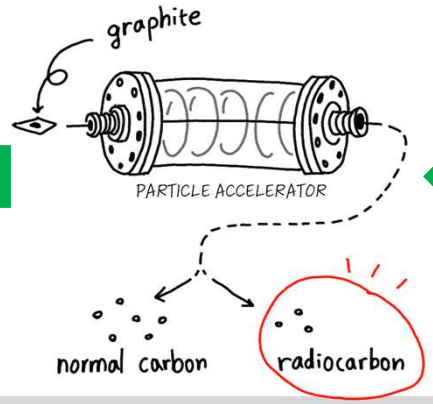
2. Wash the sample with acid.



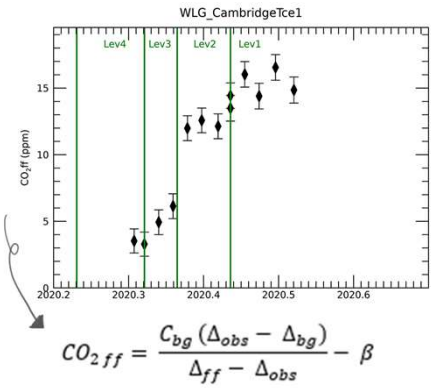
3. Burn the grass to collect CO_2.



4. Combine the CO_2 with hydrogen gas to create graphite



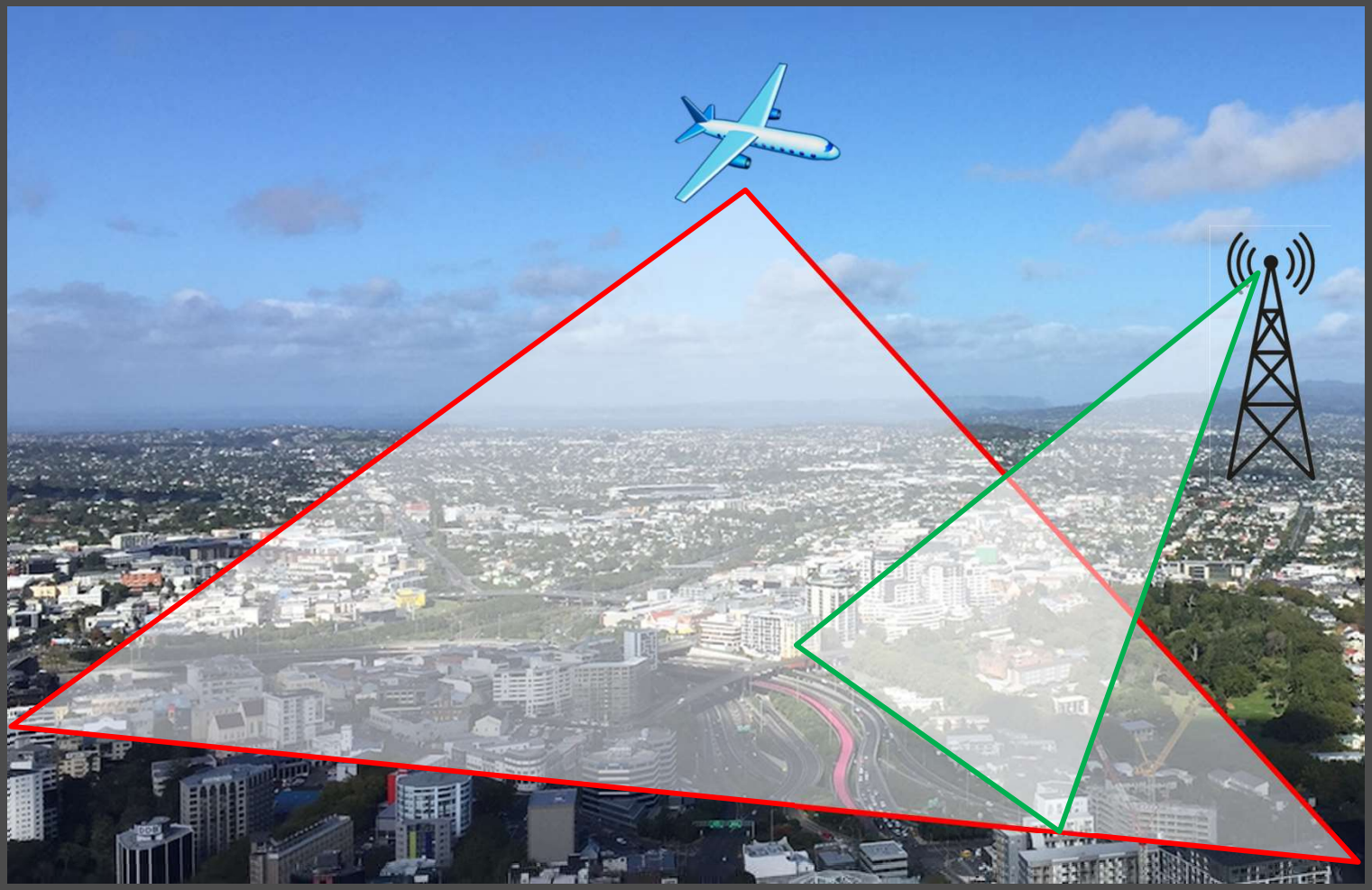
5. Use the particle accelerator to separate radiocarbon.



6. Use maths to find out how much fossil fuel CO_2 was in the air.

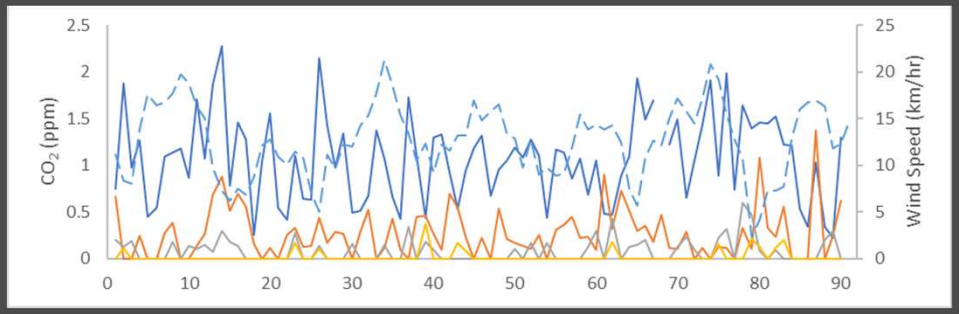
Spatial extent of sampling methods

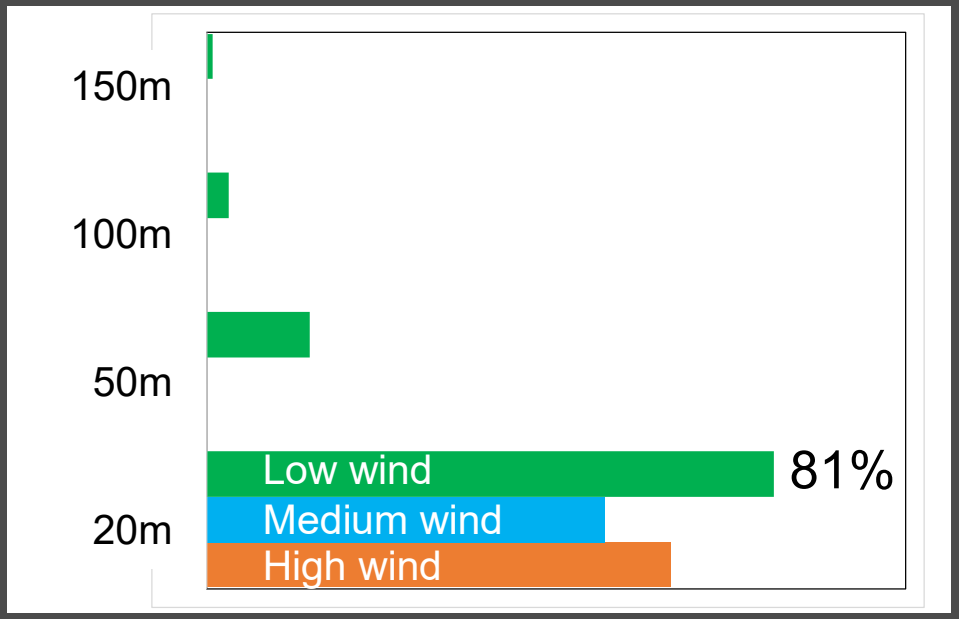
Spatial extent of sampling methods



Aircraft at high altitude and/or downwind have a footprint that covers all/most of a city
Towers can see all or part of a city depending on location and height

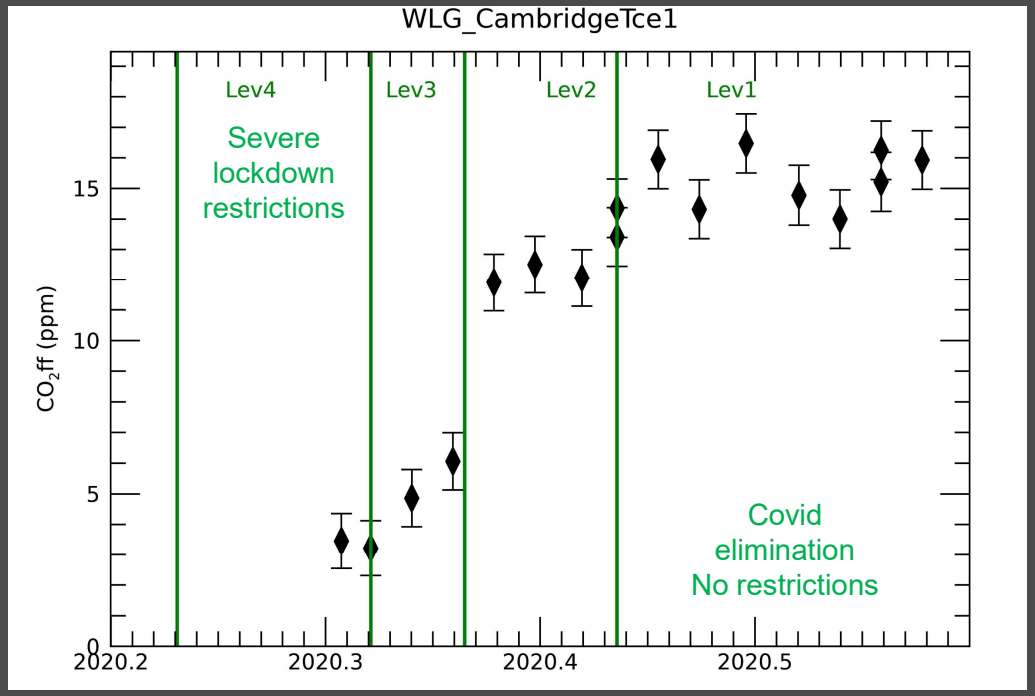
Spatial extent of sampling methods





For low growing grass is most strongly influenced by emissions within 20 m
Stronger wind reduces overall atmospheric mixing ratio
= Roadside samples see local traffic

Wellington roadside grass samples

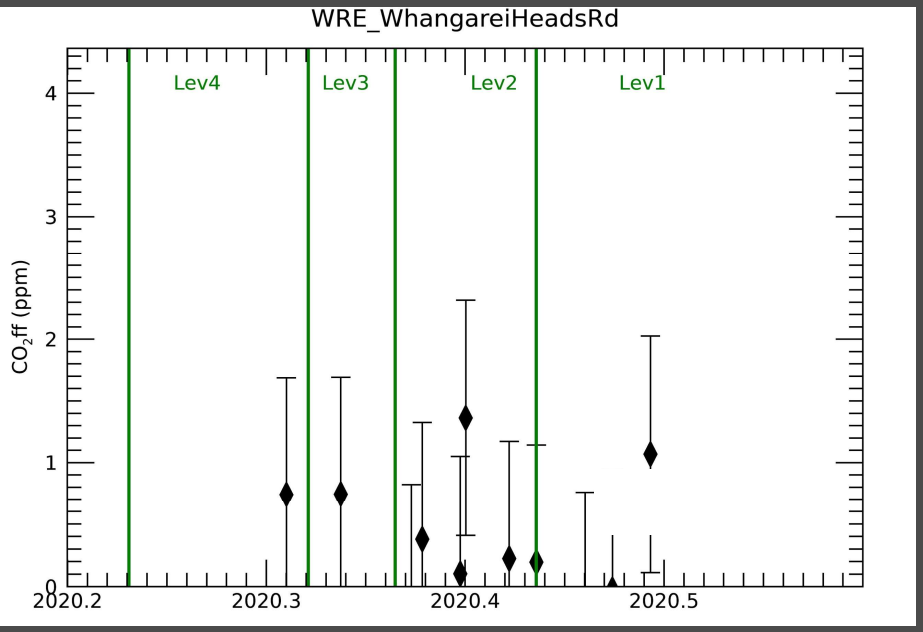


Clear change in CO_2ff concentration from Level 4 to Level 1

Cannot evaluate absolute emissions

Can evaluate the change in emissions

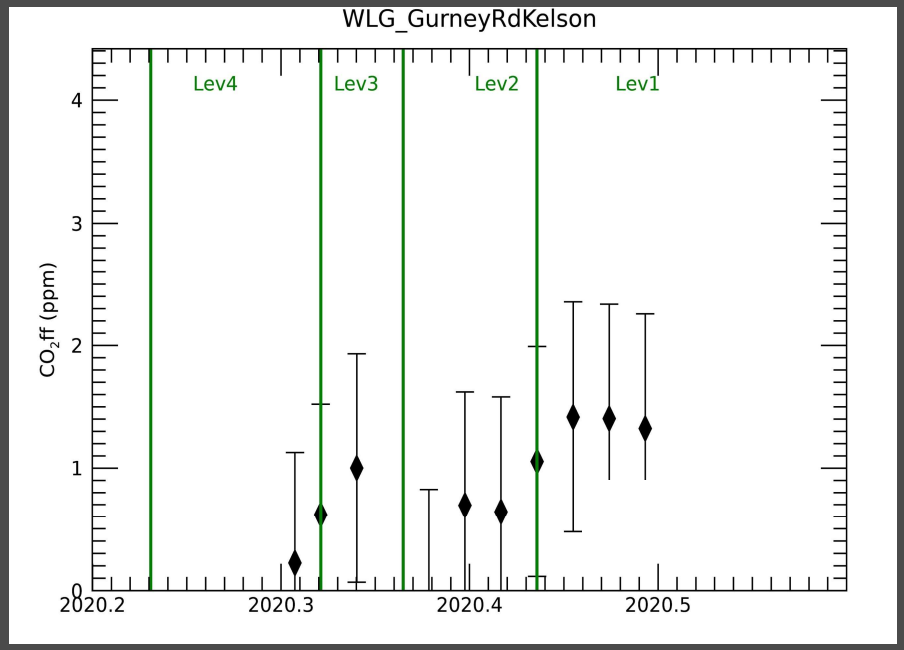
Not all sites give usable results
Clean air sites



No substantial local CO_2ff source

Still useful to diagnose “background”

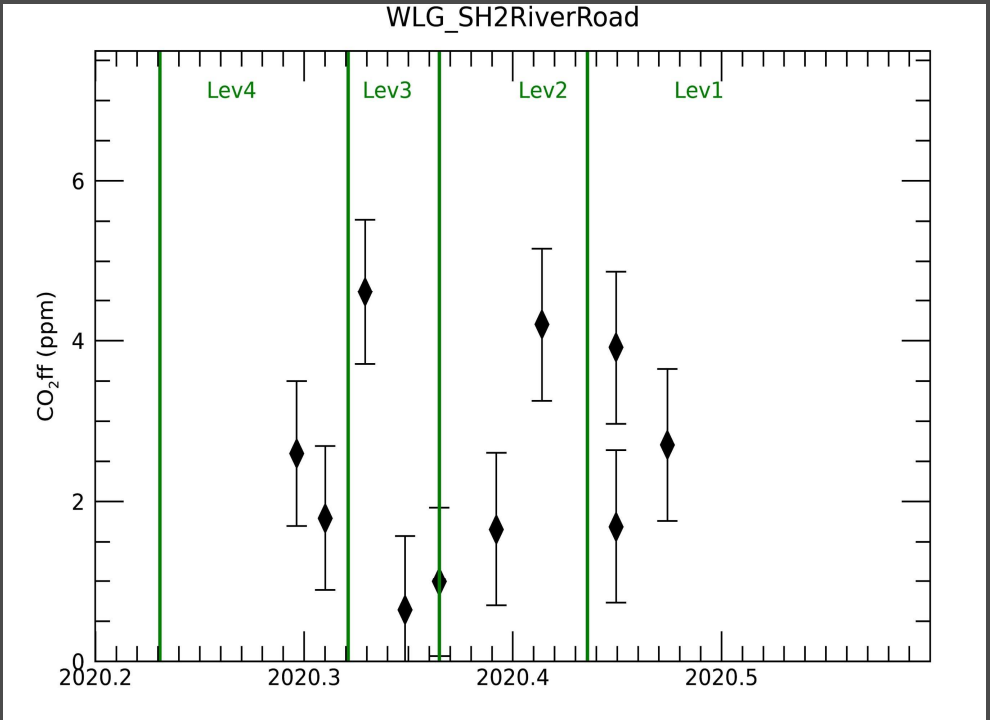
Not all sites give usable results
Sites further from large sources

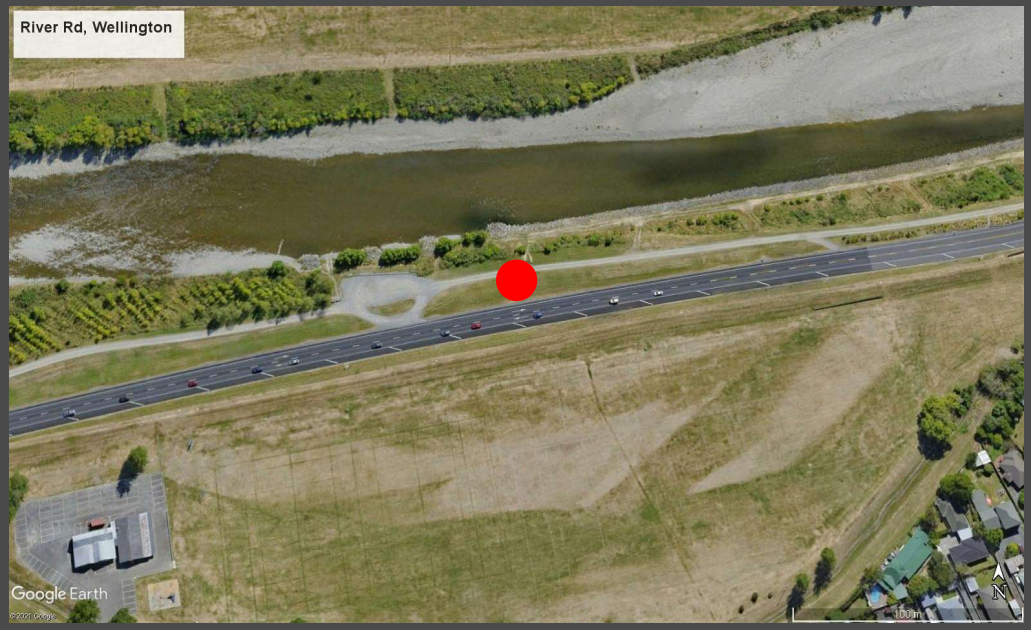


CO_2ff signal observed but small

Uncertainties too large to be meaningful

Not all sites give usable results
Sites with high variability

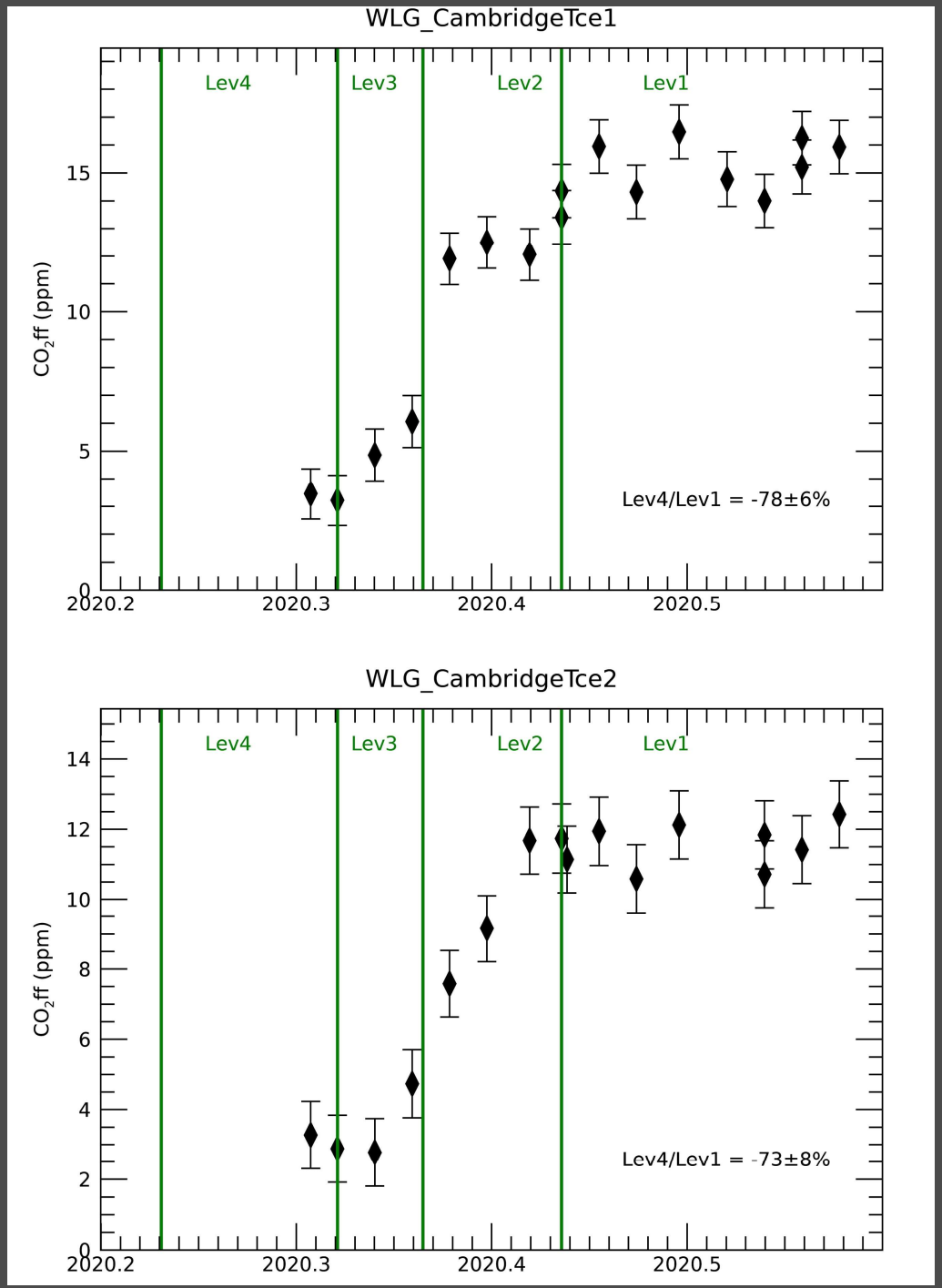




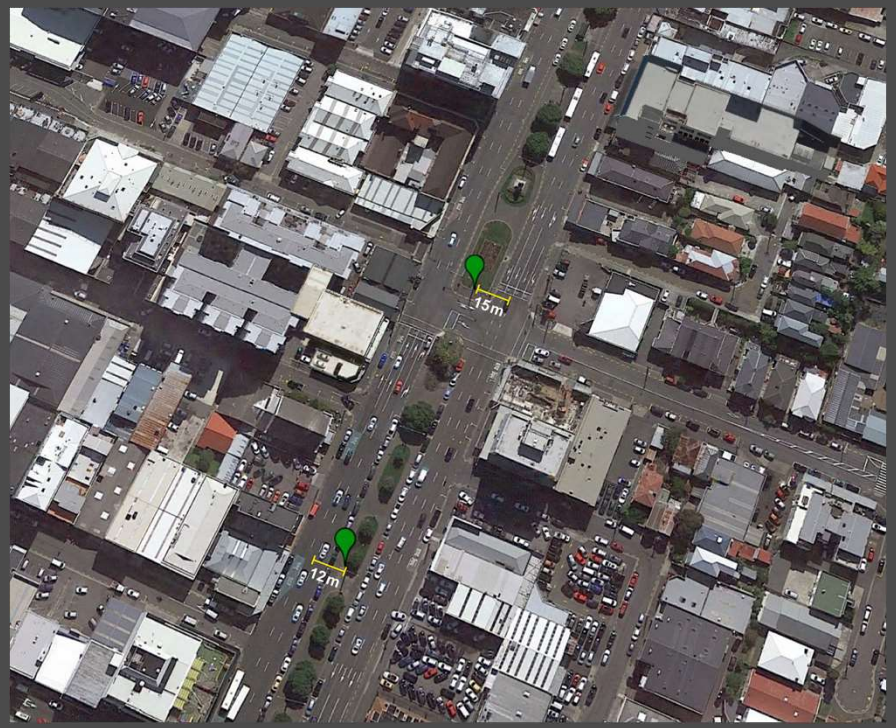
High week-to-week variability even during Level 1 when traffic emissions should be roughly constant

Meteorological variability and/or intermittent sources dominate week-to-week variability

Exclude sites with high variability in Level 1 from analysis

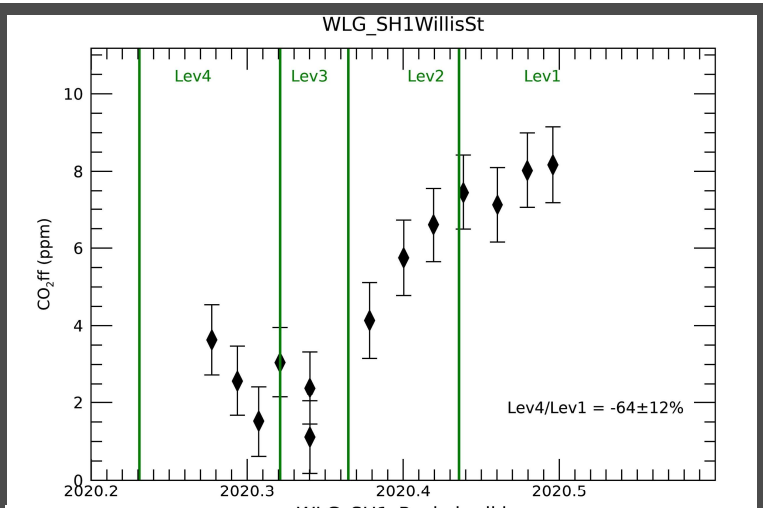


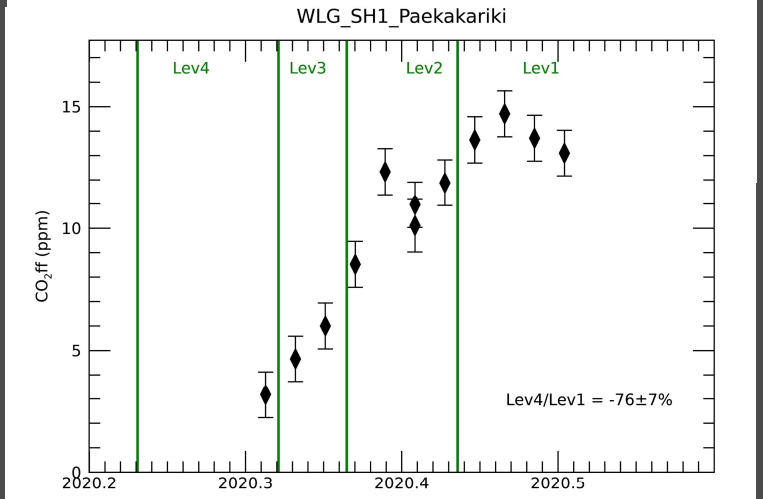
Wellington roadside grass samples

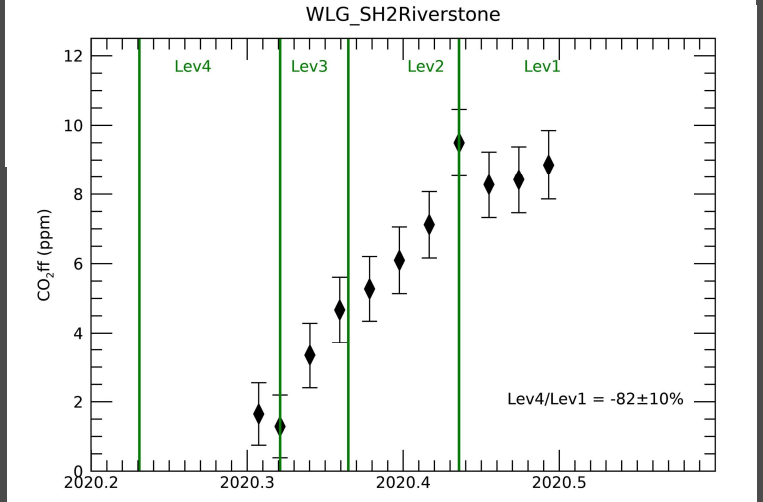


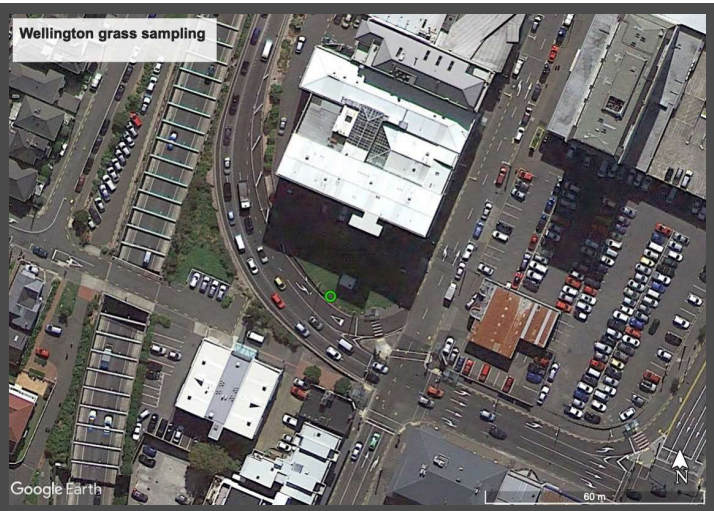
Clear change in CO_2ff concentration from Level 4 to Level 1

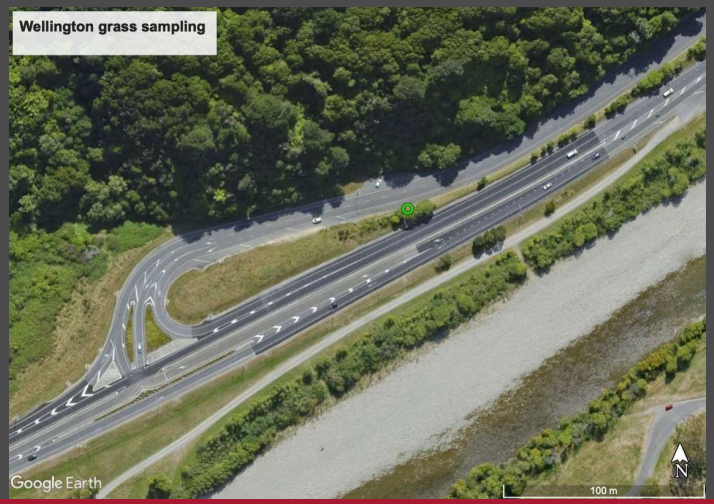
Subtle differences depending on location





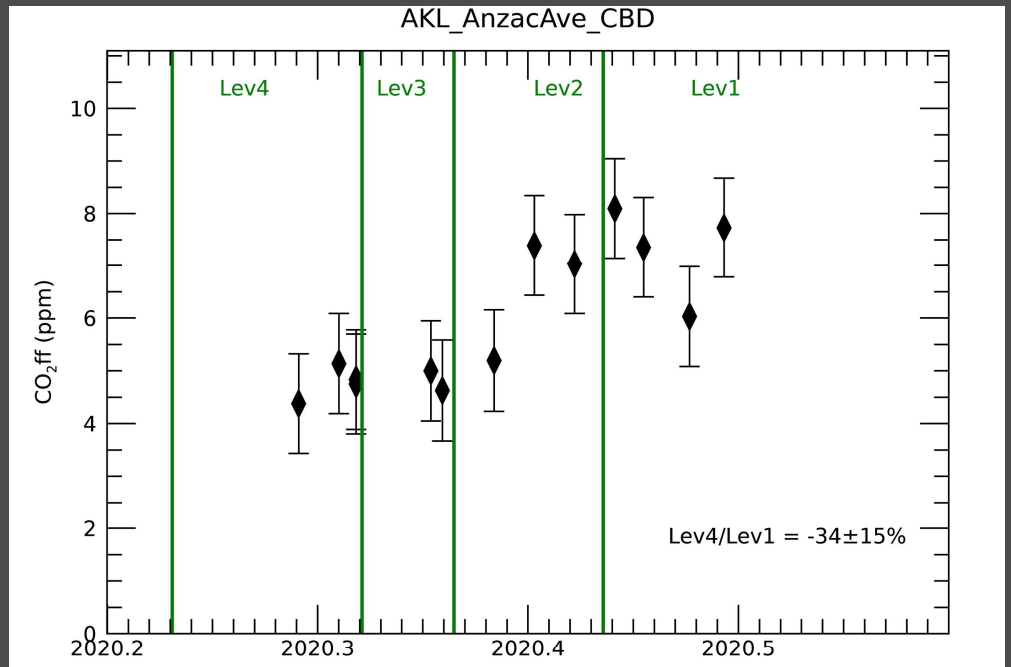


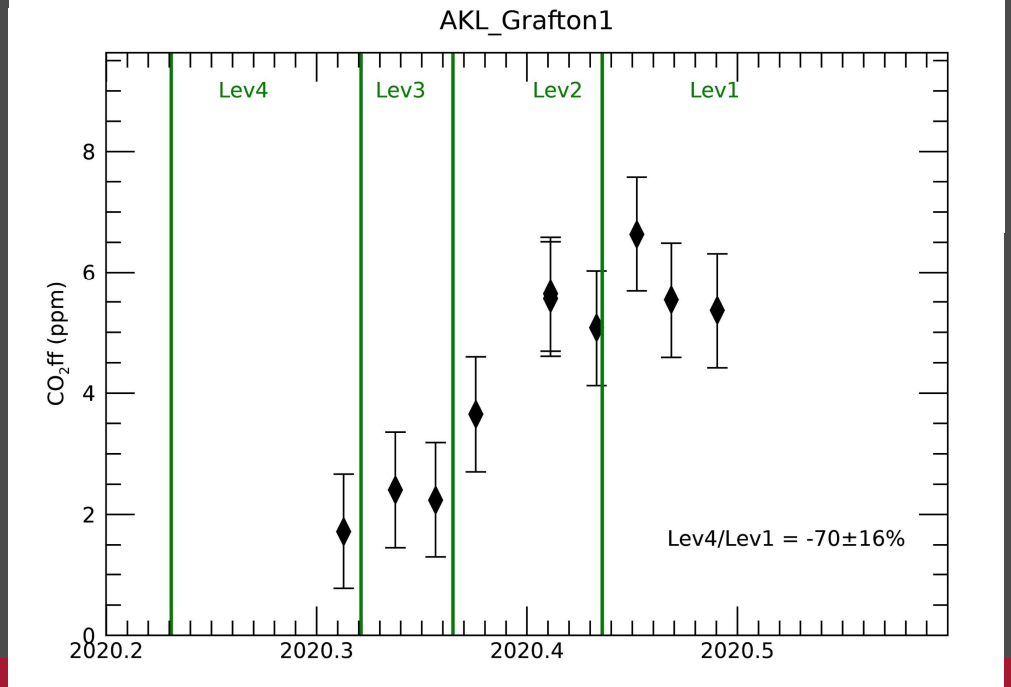




Wellington roadside grass samples

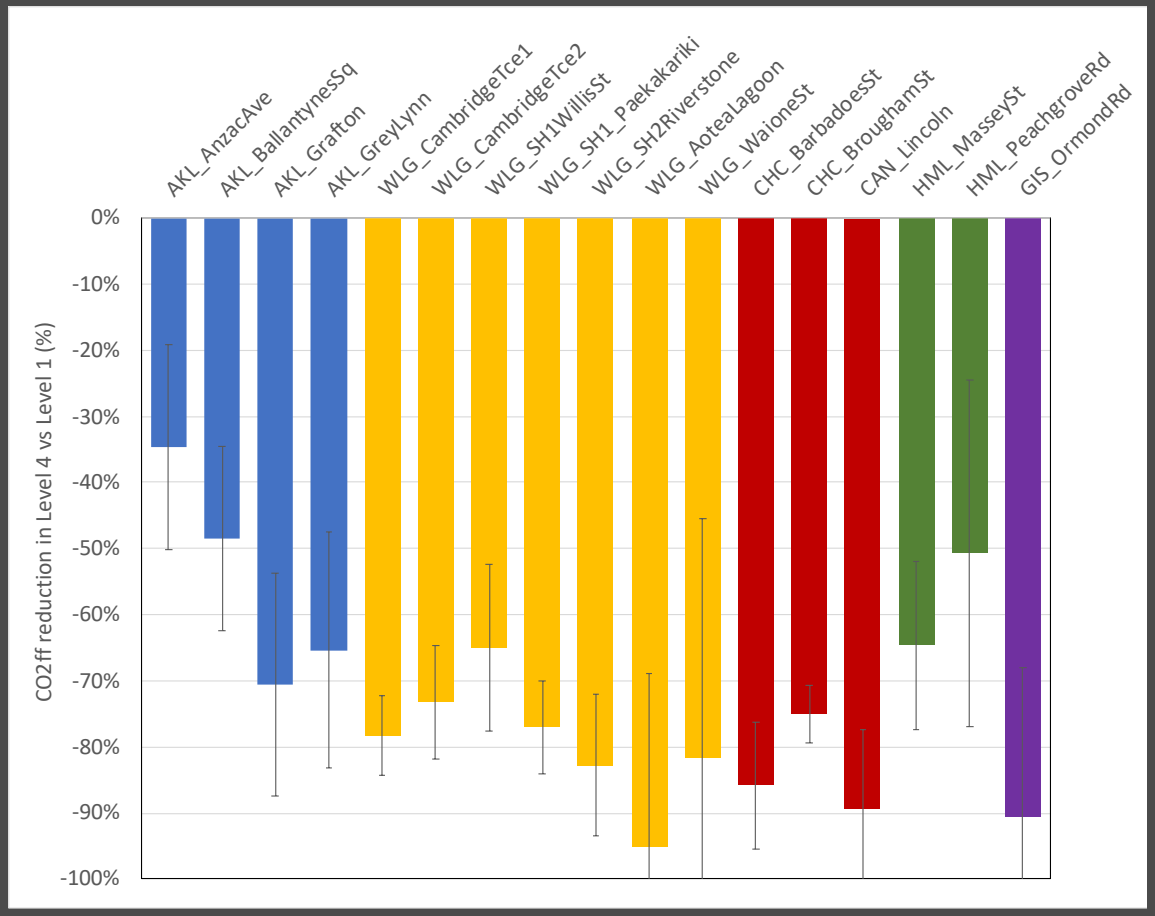
Auckland grass samples

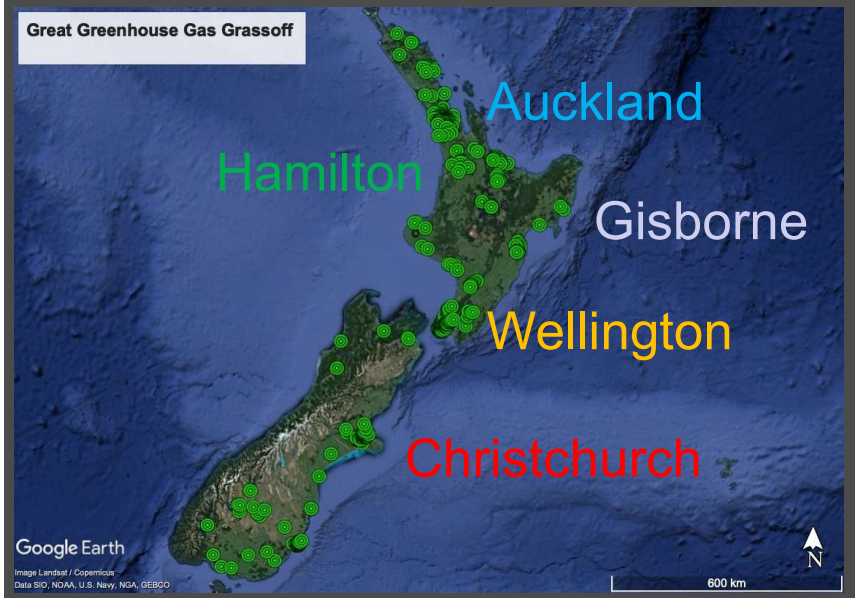




Much smaller emission drop at central Auckland site

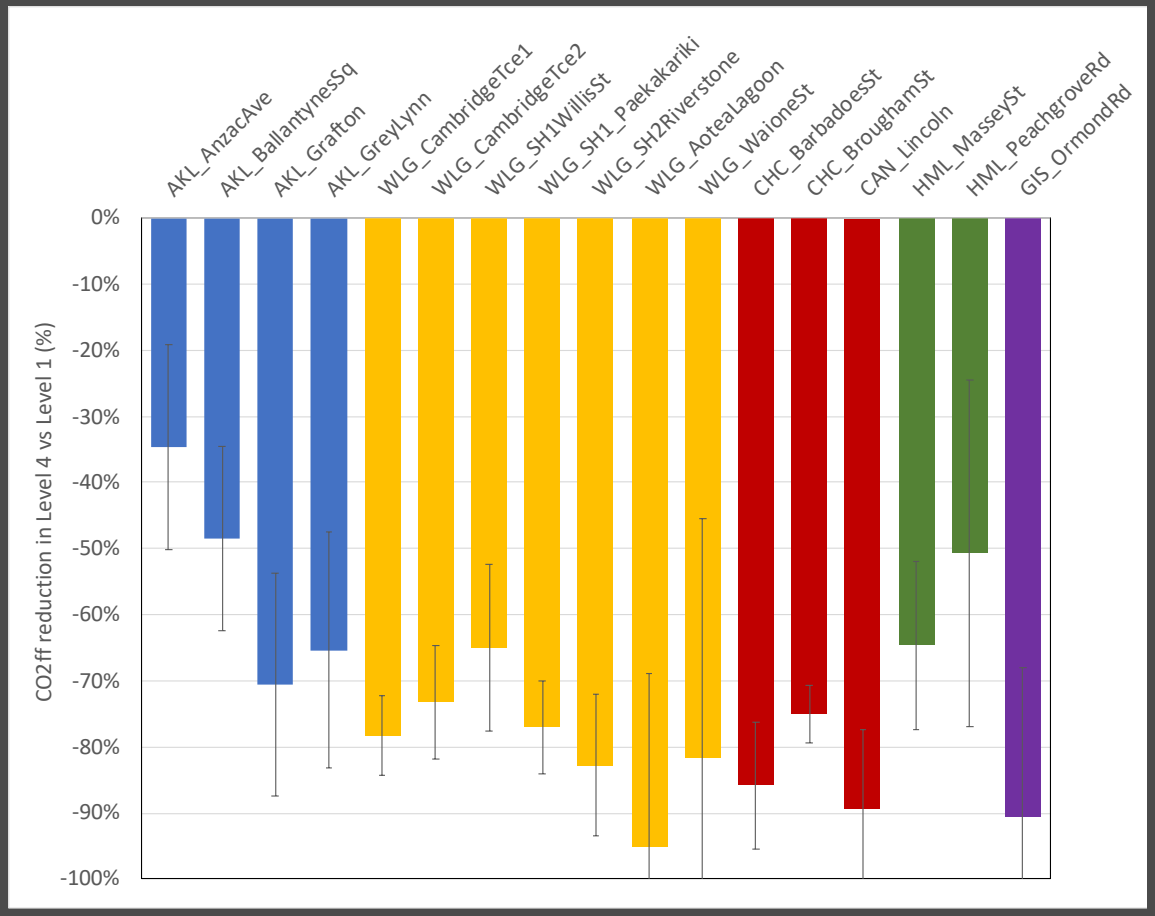
Observed Changes in CO_2ff Emissions

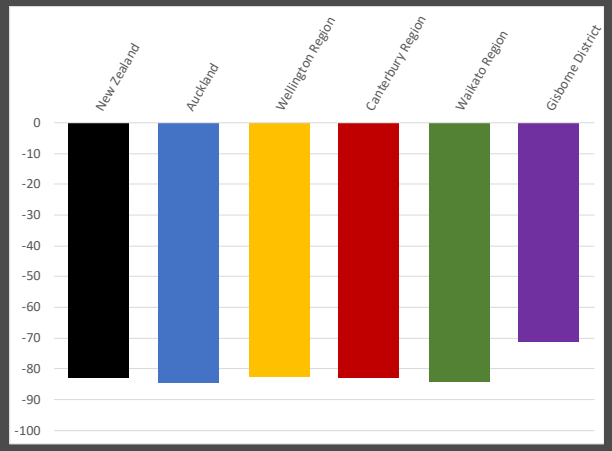




Our atmospheric observations
-75% average drop in CO_2ff during Level 4
Central Auckland sites have smallest emission decreases

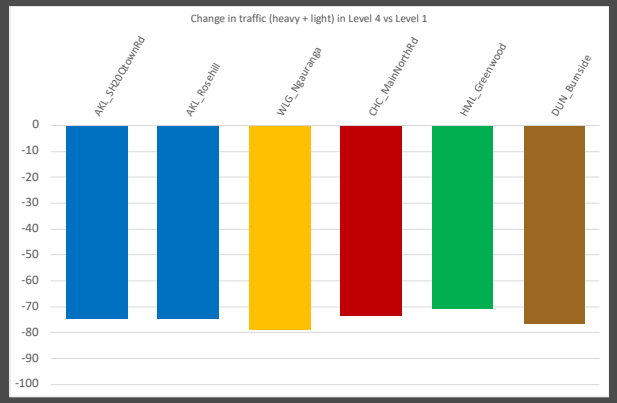
Observed Changes in CO_2ff Emissions





Apple mobility data

Typically overestimates the drop in emissions
-81%



Waka Kotahi/NZ Transport Authority traffic counts

Consistent with observed CO_2ff
-75%
But less spatial information available

Our atmospheric observations
-75% average change in CO_2ff during Level 4
Central Auckland sites have smallest emission decreases

Conclusions and ongoing work

Radiocarbon is readily used to determine recently added fossil fuel CO_2 into the atmosphere

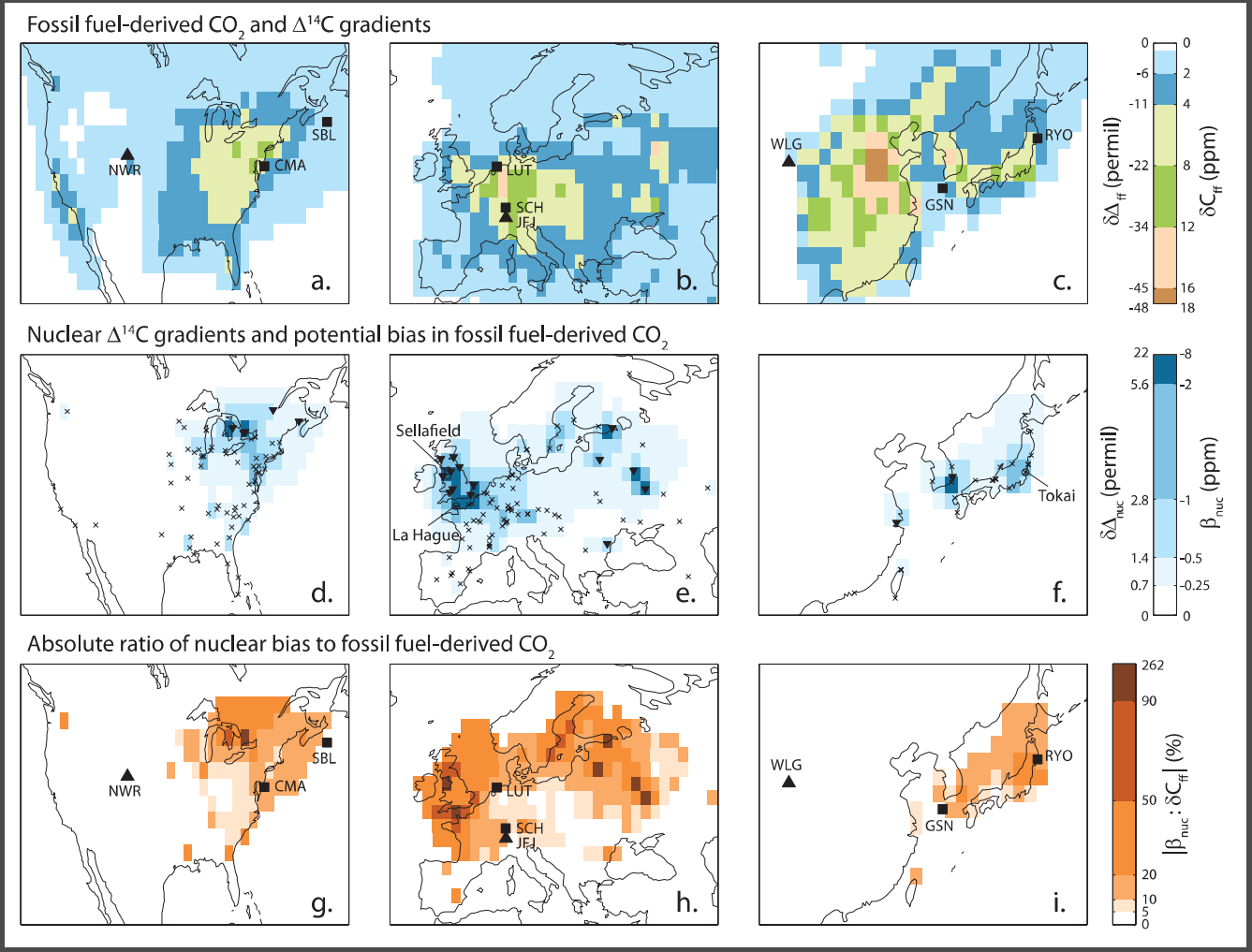
Emission rates for urban and other areas can be evaluated to improve emission reporting and mitigation action

Traffic emission decreases of 75% during Level 4 lockdown from 17 sites across 5 New Zealand cities, consistent with traffic data

-suggests that future decoupling of traffic and emissions could be readily tracked with ^{14}C

Jocelyn Turnbull j.turnbull@gns.cri.nz

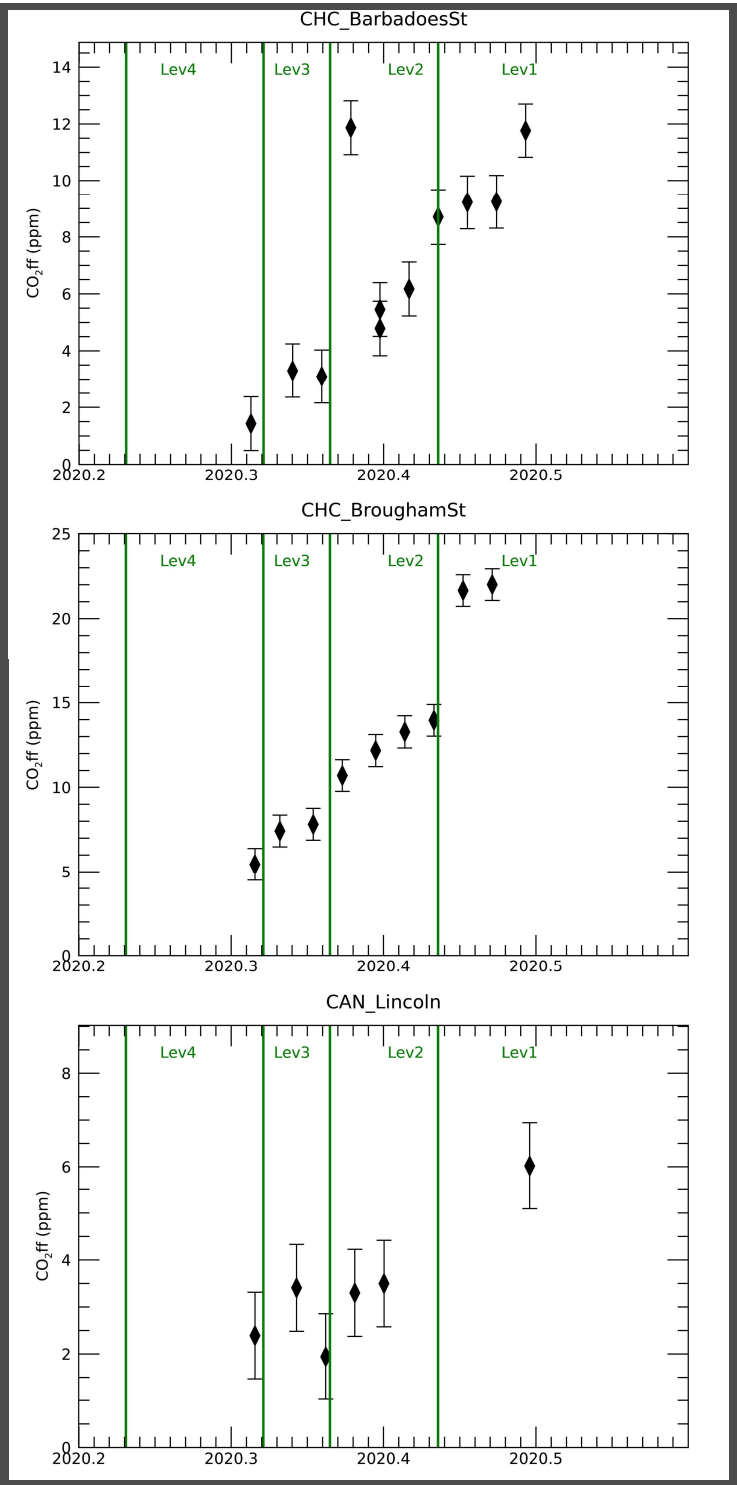
Nuclear bias

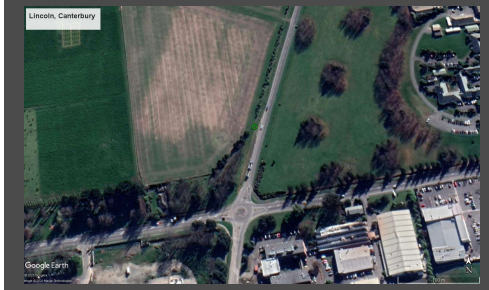


Graven and Gruber, 2011

^{14}C emitted from some types of nuclear power plant can cause an underestimate of CO_2ff

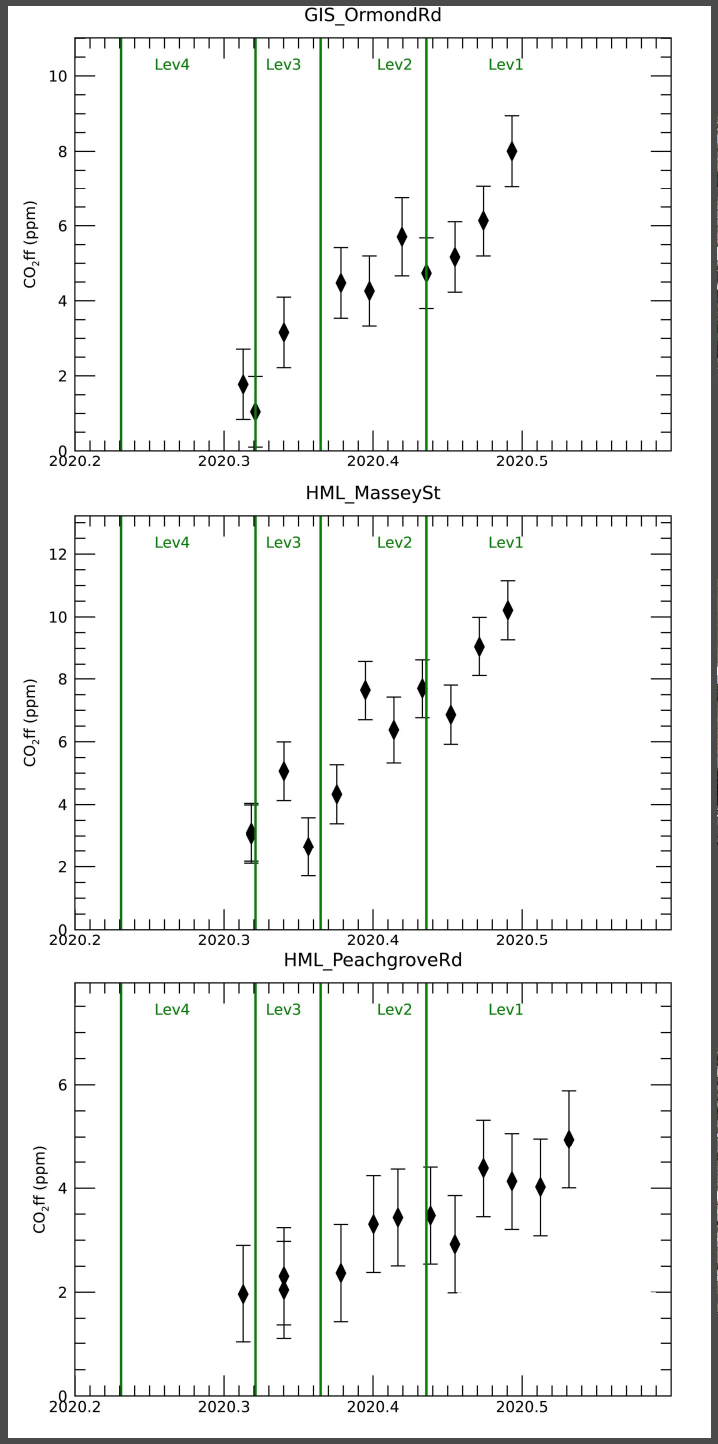
Christchurch Grass Sampling

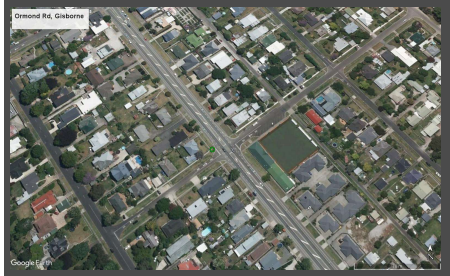


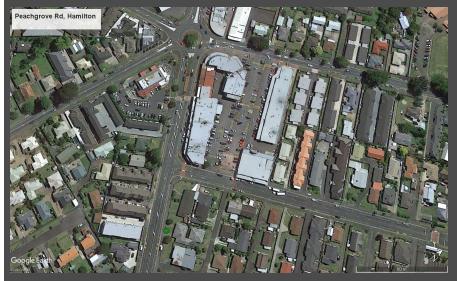


Similar to results for Wellington

Rural Canterbury road shows more modest change







Hamilton and Gisborne

Two sites show 70-80% emission drops, similar to Wellington and Christchurch

Peachgrove Rd Hamilton shows more modest drop in emissions during Level 4