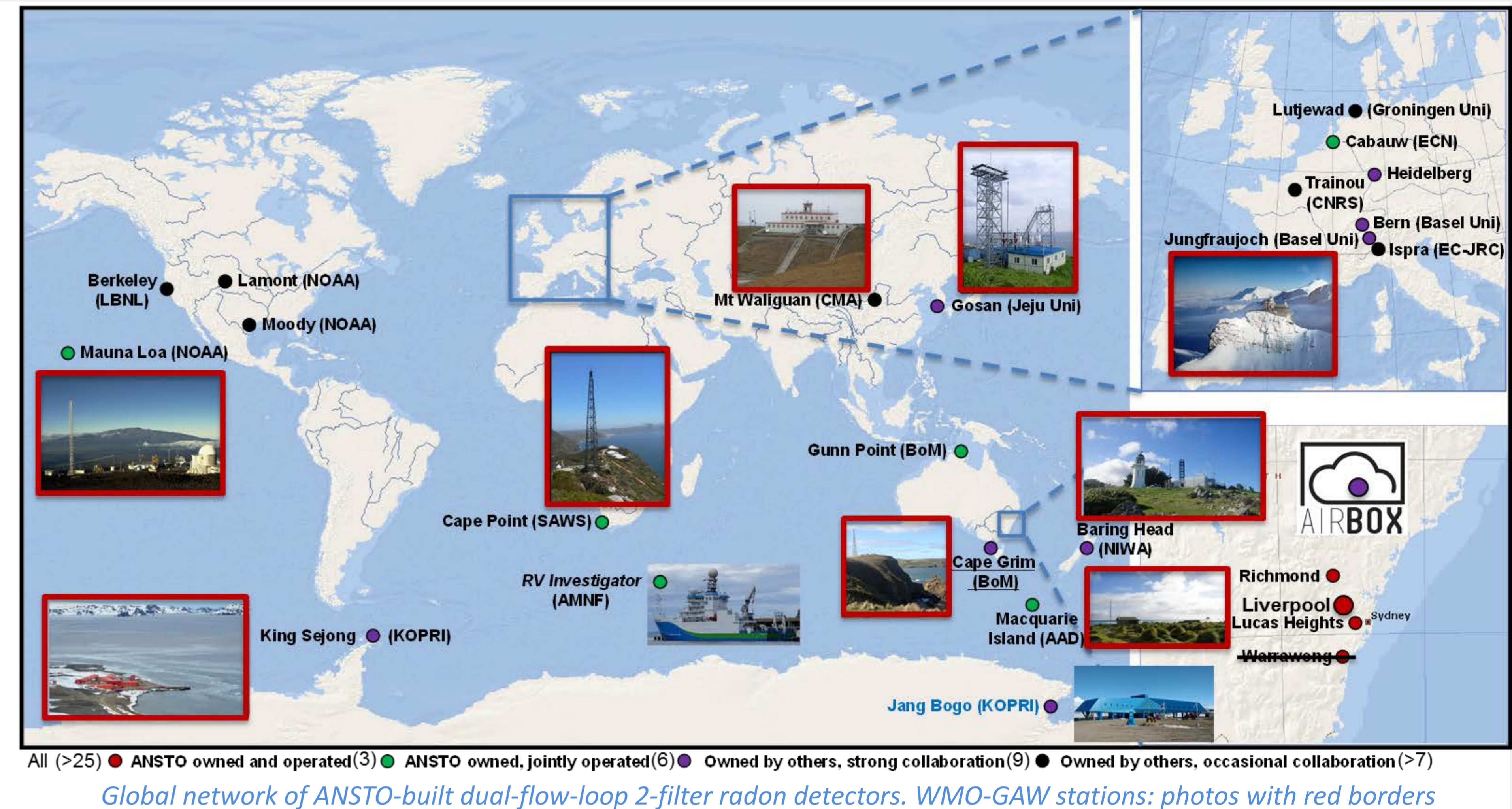


Radon-222 provides an unambiguous indication of the degree of recent terrestrial influences on an air mass. This makes it an ideal tracer at baseline, high-elevation and remote stations, where it is used for a range of applications including: interpretation of changes in atmospheric composition / baseline; tracking of movements in greenhouse gases and other trace species; calibration of regional emissions; and evaluation of regional and global transport models.

The radon program at the *Cape Grim Baseline Air Pollution Station* in Tasmania, operated jointly by the Australian Nuclear Science and Technology Organisation (ANSTO) and the Australian Bureau of Meteorology, is the premier atmospheric radon monitoring program worldwide. The dual-flow-loop two-filter detector design pioneered at ANSTO is recognised by WMO-GAW as providing the international benchmark in radon monitoring for global and regional atmospheric composition studies.

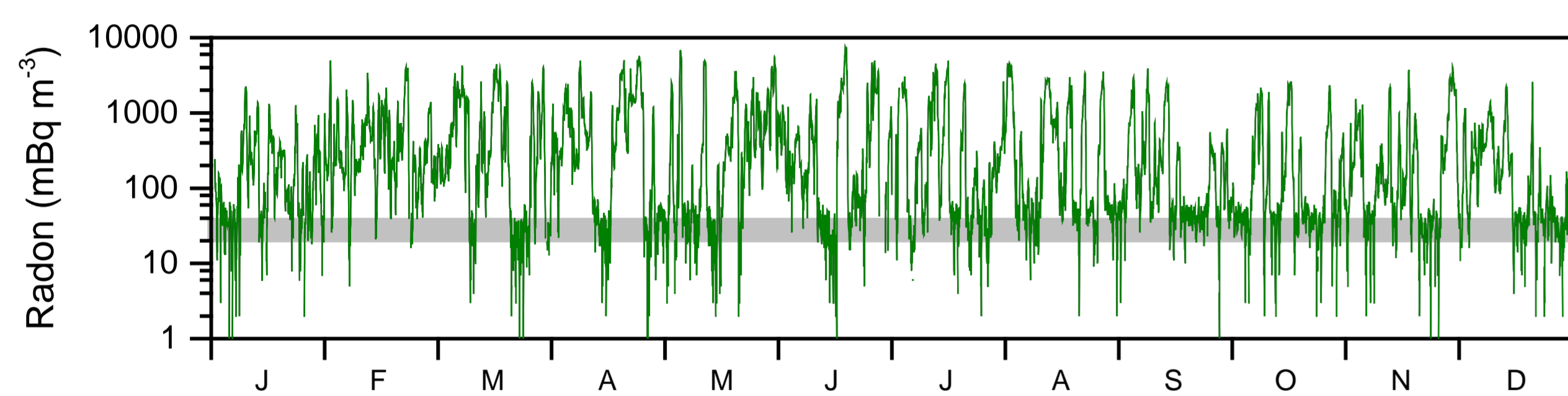
By forming and maintaining partnerships with leading international players such as NOAA/ESRL, ANSTO has been able to collaboratively operate a number of long-term radon measurement programs at other stations in the WMO-GAW network. Multi-year radon data sets are available from Mauna Loa Observatory in Hawaii, Cape Point in South Africa, Gosan in Korea and Jungfraujoch in the Swiss Alps.

In recent years, there has also been an effort to “fill gaps” in the coverage of radon observations in data-poor regions of the Southern Hemisphere, by establishing new measurement programs at existing and newly-established GAW stations. Collaborations with the Australian Antarctic Division, CSIRO and the Korean Polar Research Institute have led to the establishment of radon programs at Macquarie Island, aboard the *RV Investigator*, and two sites on the Antarctic coastline.



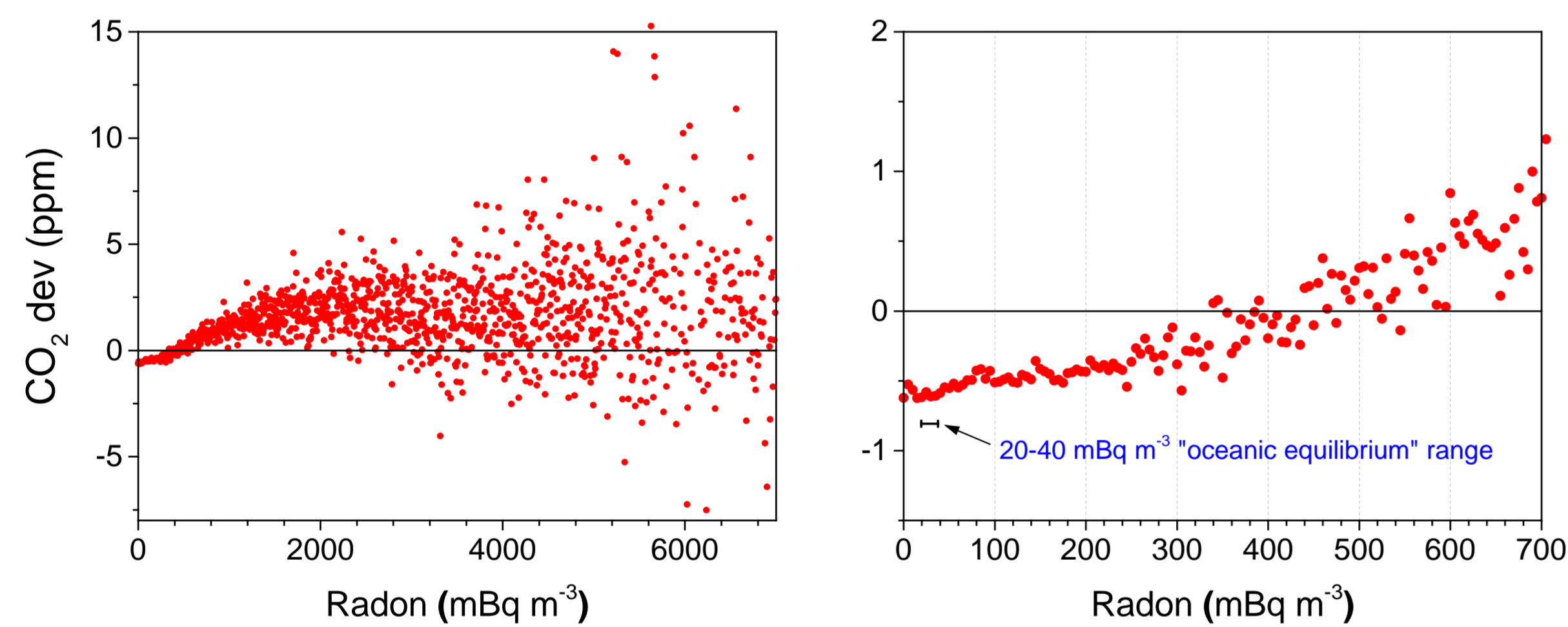
Redefining “baseline” conditions

Since radon (^{222}Rn) has an almost exclusively terrestrial source, radon measurements are often used as a proxy for terrestrial influence, or “pollution potential”, of an air mass.



Hourly ^{222}Rn measurements at Cape Grim in 2010 indicating a wide range of terrestrial influence, from recent (2-6 Bq m^{-3}) to baseline oceanic (0.02 - 0.04 Bq m^{-3} , shaded in grey). Note the logarithmic scale.

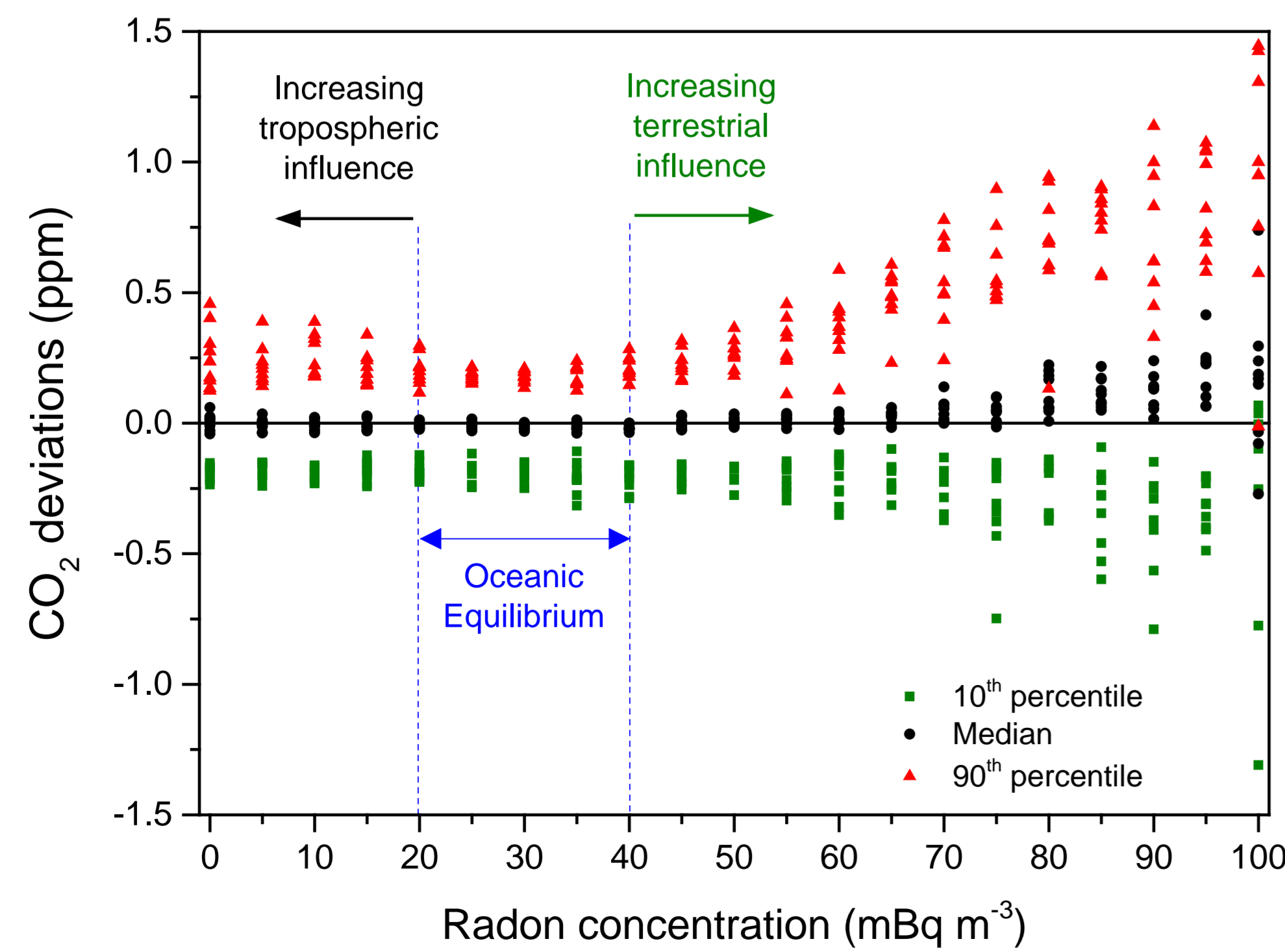
Concentrations of atmospheric trace species, like carbon dioxide (CO_2), can be very sensitive to the degree of recent terrestrial influence.



CO_2 mixing ratio deviations (from the monthly mean value) as a function of increasing terrestrial influence (^{222}Rn). With increasing time away from land (decreasing ^{222}Rn) both positive (source) and negative (uptake) deviations from the monthly mean converge towards a consistent value representative of baseline marine conditions.

Traditionally, baseline conditions at Cape Grim have been selected based on a combination of parameters, including wind direction, wind speed, particle number and signal stability, in conjunction with advanced filtering, data outlier exclusion strategies and a simple radon threshold.

The accuracy and performance of early ^{222}Rn detectors necessitated a relaxed threshold of 100 mBq m^{-3} for significant terrestrial influence. However, for over a decade the 3-10 mBq m^{-3} LLD of modern detectors has enabled a more detailed examination of the characteristics of air masses with $^{222}\text{Rn} < 100 \text{ mBq m}^{-3}$ (Chambers et al. 2016).



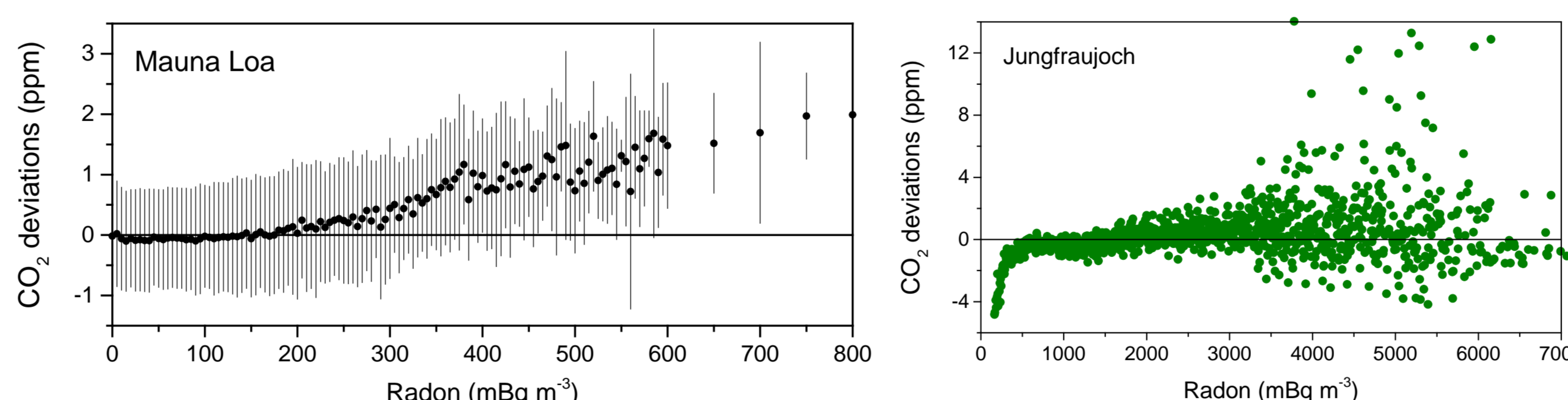
Distributions of CO_2 mixing ratio deviations are narrowest within the range of ^{222}Rn concentrations for air masses in oceanic equilibrium (20-40 mBq m^{-3}). Outside of this range, air masses show signs of increasing terrestrial, or tropospheric, influence.

A universal baseline tool for contrasting sites

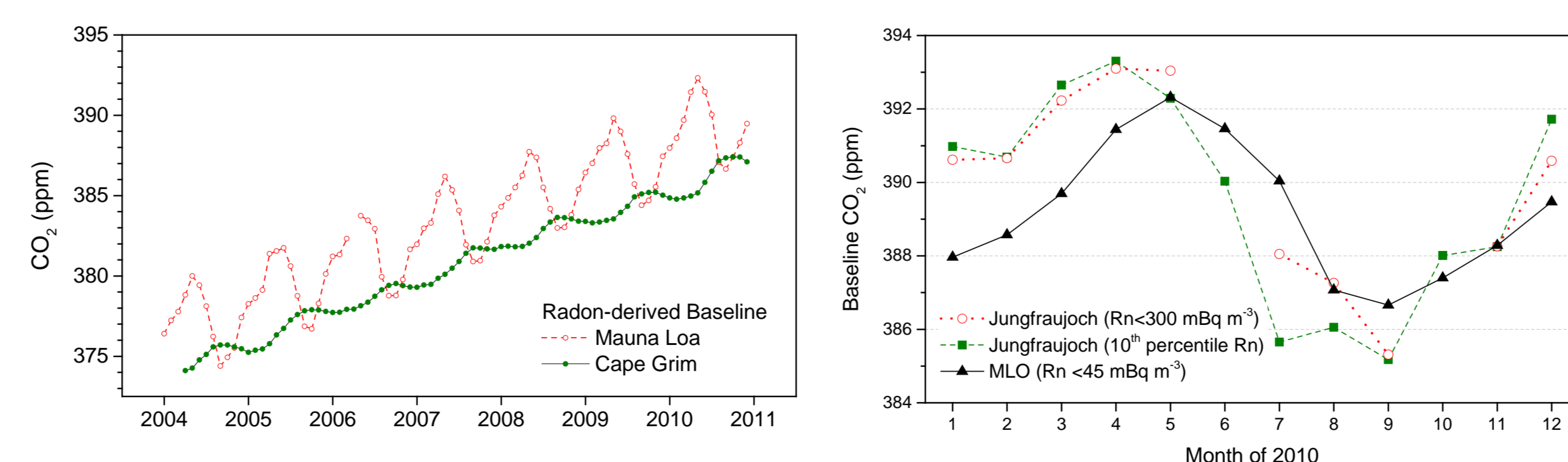
For sufficiently sensitive modern detectors (1500 and 5000 L dual-flow-loop two-filter models) radon observations *alone* can provide a consistent and universal (site independent) method for unambiguous baseline identification. Furthermore, for continental sites with complex topography and meteorology, where true “baseline” conditions are rarely encountered, radon can be used to identify the least terrestrially-perturbed air masses, and provide an objective means by which to apply limits to the level of “acceptable terrestrial influence” for a given application.



The collection of World Meteorological Organisation Global Atmosphere Watch baseline stations (8 of which employ ANSTO radon detectors) span a wide range of physical settings including: remote oceanic, coastal, high-elevation island and continental sites, and polar regions, each with specific baseline selection requirements.

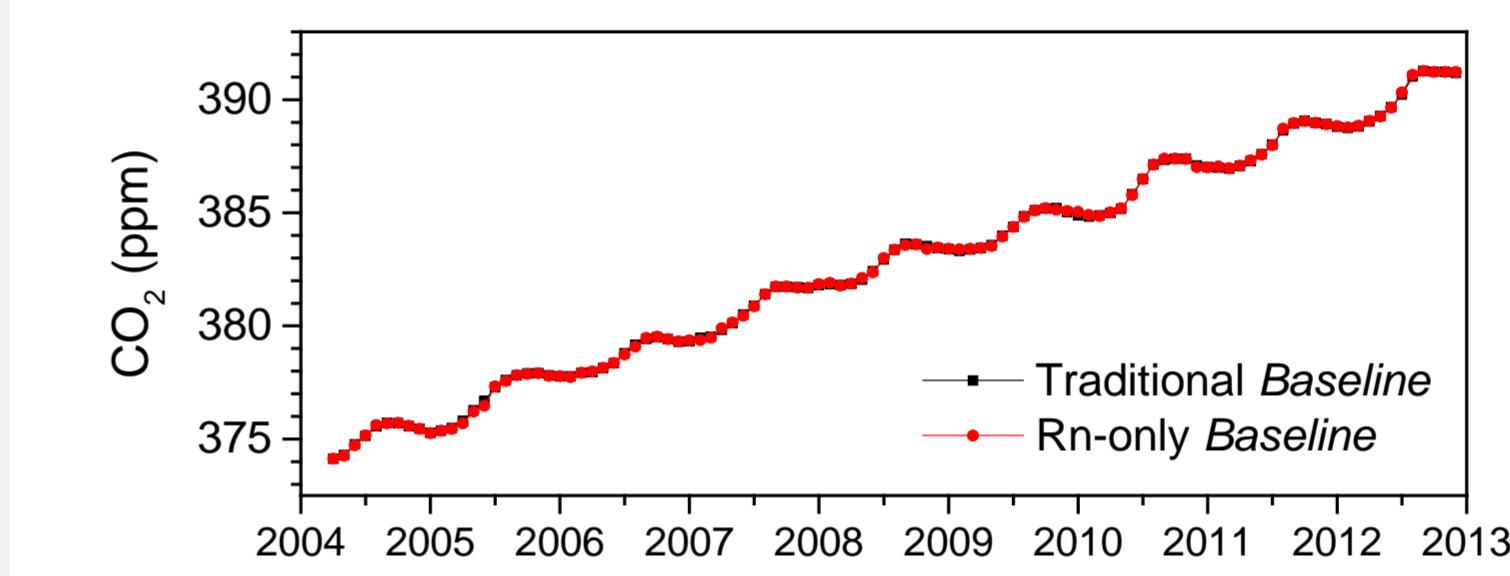
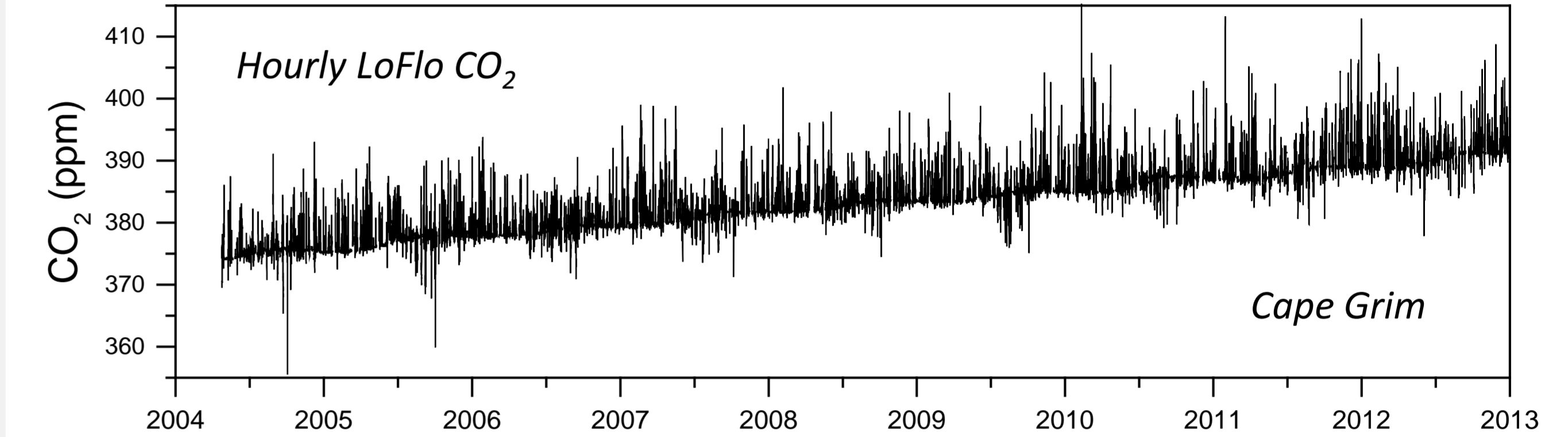


At the remote island high-altitude Mauna Loa site CO_2 converges to a consistent value at ^{222}Rn values $< 150 \text{ mBq m}^{-3}$. At the high-altitude continental site Jungfraujoch, however, no such CO_2 baseline plateau is observed. In this case, ^{222}Rn can be used to define an objective limit for air mass terrestrial influence for a “continental background”.

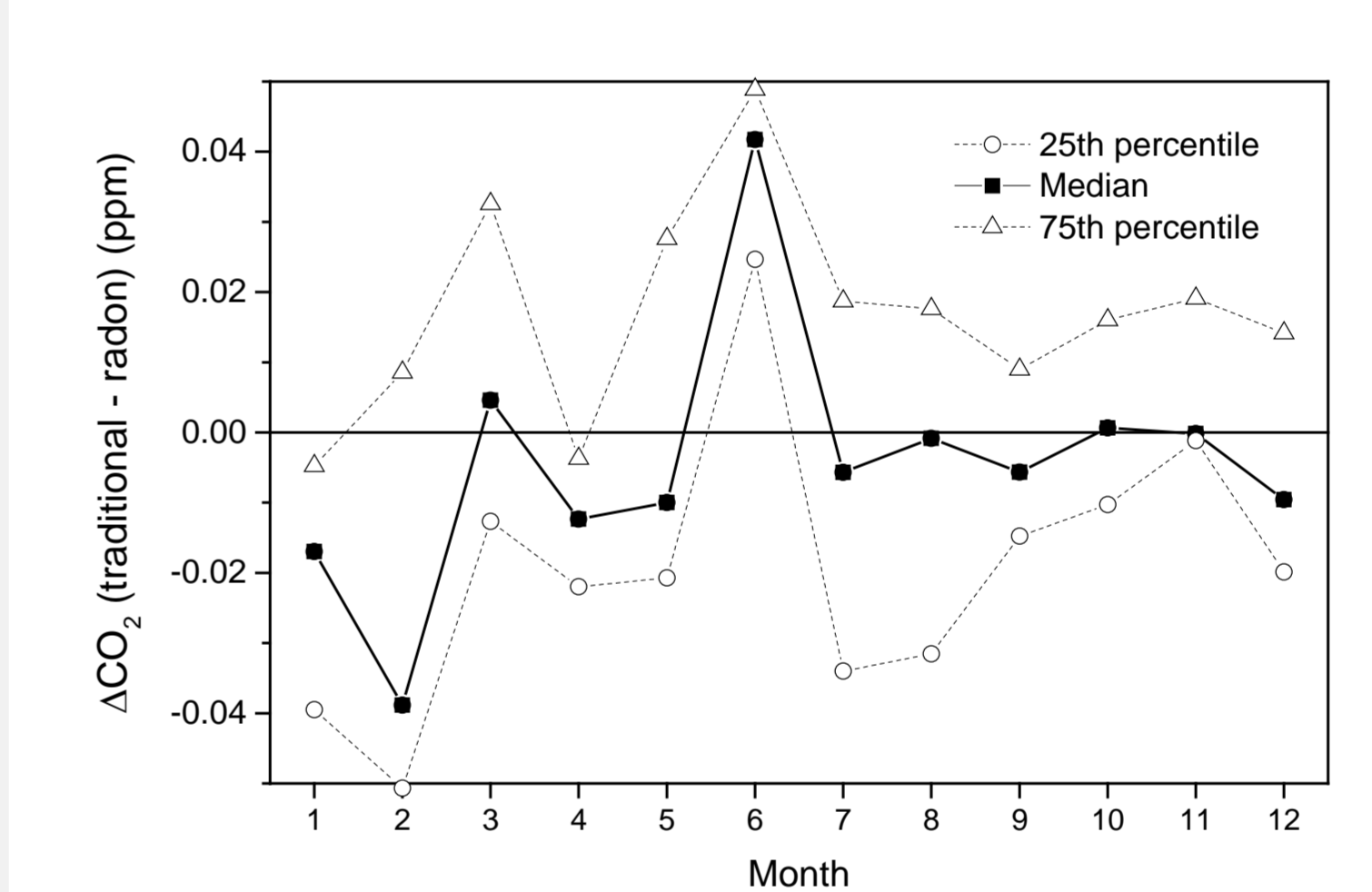


Rn-derived baseline CO_2 signal at Cape Grim and Mauna Loa, and a comparison of the monthly CO_2 “continental background” at Jungfraujoch with MLO baseline values.

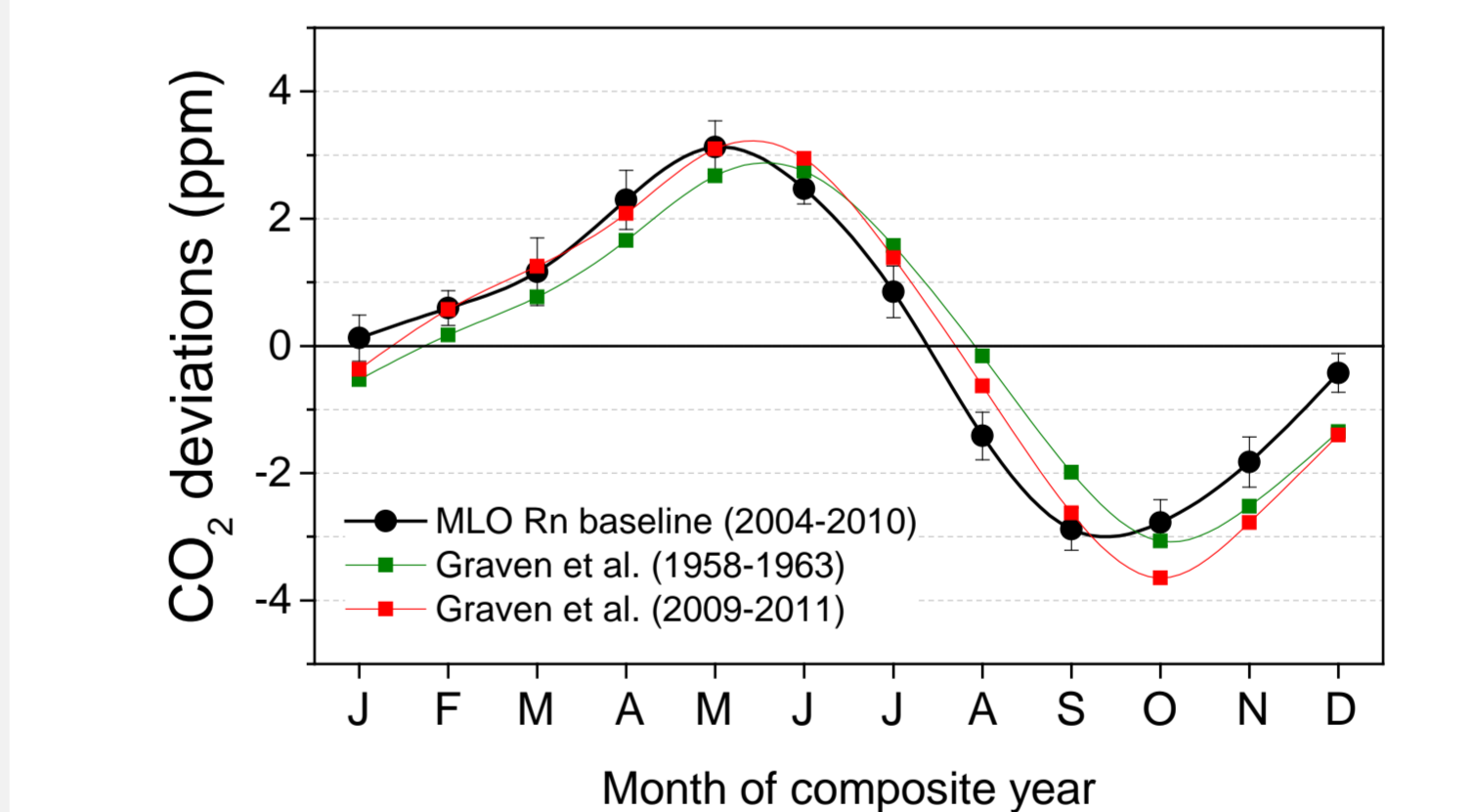
Radon vs traditional baseline techniques



On first inspection, baseline CO_2 derived from traditional (wind speed & direction, signal stability, outlier removal and curve fitting) and radon-only methods are identical.



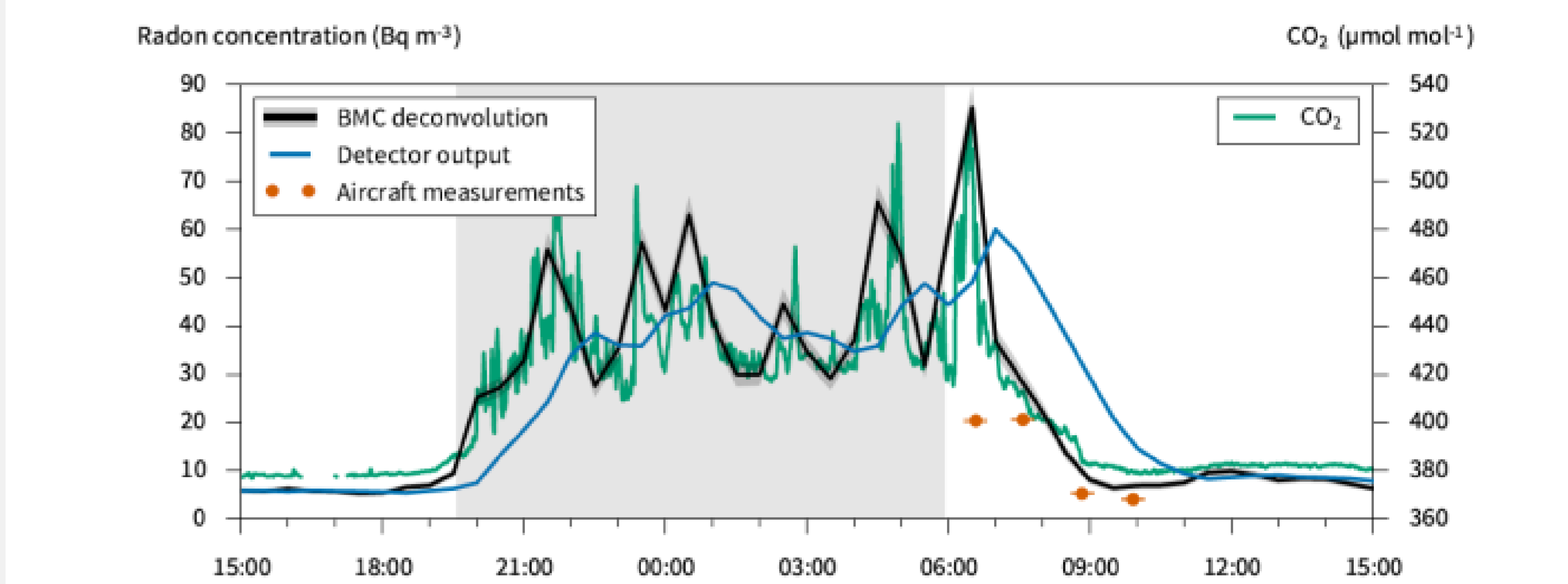
However, small but systematic differences exist between results using the two techniques.



Traditional techniques yielded lower CO_2 in summer and higher values in winter cf. the radon approach. Subsequent radon analyses linked many of these differences to vestigial terrestrial influences.

Continual improvement: response time correction

ANSTO detectors offer the lowest ^{222}Rn detection limits worldwide, but have been hampered by their 45 min response time. A deconvolution tool was recently developed (Griffiths et al. 2016) that corrects for the response time and improves the temporal resolution. This development will improve the utility of ^{222}Rn observations for evaluating transport and mixing schemes in numerical models at both baseline and inland sites.



A diurnal cycle of near-surface hourly ^{222}Rn observations (with and without the deconvolution response time correction) compared with high-resolution CO_2 observations and aircraft “grab-samples” of ^{222}Rn from 20 m agl.

Chambers, S.D., et al. Towards a universal “baseline” characterisation of air masses for high- and low-altitude observing stations using Radon-222. *Aerosol and Air Quality Research*, 16, 885–899, 2016.
 Graven, H.D., et al. Enhanced Seasonal Exchange of CO_2 by Northern Ecosystems Since 1960. *Science*, 341: 1085, 2013.
 Griffiths, A.D., et al. Increasing the accuracy and temporal resolution of two-filter Radon-222 measurements by correcting for the instrument response. *Atmospheric Measurement Techniques*, 9, 2689–2707, 2016.