# Characterising diurnal & synoptic timescale changes in urban air quality using Radon-222

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**EGU General Assembly 2020**

**GI6.1 Geoscience applications of environmental radioactivity**

3 - 8 May 2020
Vienna, Austria

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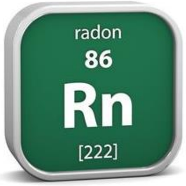
# Radon-222: friend or foe




- Indoor/underground $^{222}Rn$ activity concentrations can easily be 2-3 orders of magnitude greater than outdoor, environmental values
- Consequently, $^{222}Rn$ investigations in confined spaces focus mostly on its detrimental health effects, whereas studies of ambient outdoor $^{222}Rn$ focus more on its capabilities as an atmospheric tracer of transport and mixing
- $^{222}Rn$ is naturally-occurring, unreactive decay product of $^{238}U$ (ubiquitous in soils and rocks), so it has a *relatively* consistent source function for unsaturated/unfrozen soils
- Radon's solubility is such that – on timescales of rain movement through the atmospheric boundary layer – there is negligible washout, and it does not attach to aerosols. Consequently, radon's primary sink is radioactive decay
- Unlike greenhouse gases, $^{222}Rn$ doesn't accumulate in the atmosphere on >synoptic timescales ($t_{0.5}$ 3.8d), yet it is roughly conservative over 1 night
- This unique combination of characteristics makes radon a convenient & powerful tracer for atmospheric transport and mixing studies

# $^{222}$Rn tracing of transport and mixing

- The oceanic $^{222}$Rn flux is 2-3 orders of magnitude less than terrestrial fluxes, and there are no *significant* atmospheric radon sources
- Consequently, air masses moving out over the ocean, or from the ABL to the troposphere or stratosphere, can usually be tracked (with a suitably sensitive detector) for up to 2-3 weeks (due to radon's 3.8 d half-life)
- Over land, however, radon's half-life means that ABL air masses retain a "memory" of where they have been (fetch influences) for many days
- If there is significant spatial heterogeneity of the surface $^{222}$Rn flux, radon fetch effects can confound attempts to relate changes in [$^{222}$Rn] to mixing
- Vertical gradients of [$^{222}$Rn] near the surface avoid this problem, since observations at both heights are similarly affected by fetch effects
- Consequently, near-surface vertical [$^{222}$Rn] gradients are directly linked to the net outcomes of vertical mixing processes, and serve as a proxy for the behaviour of primary pollutants with surface-based sources

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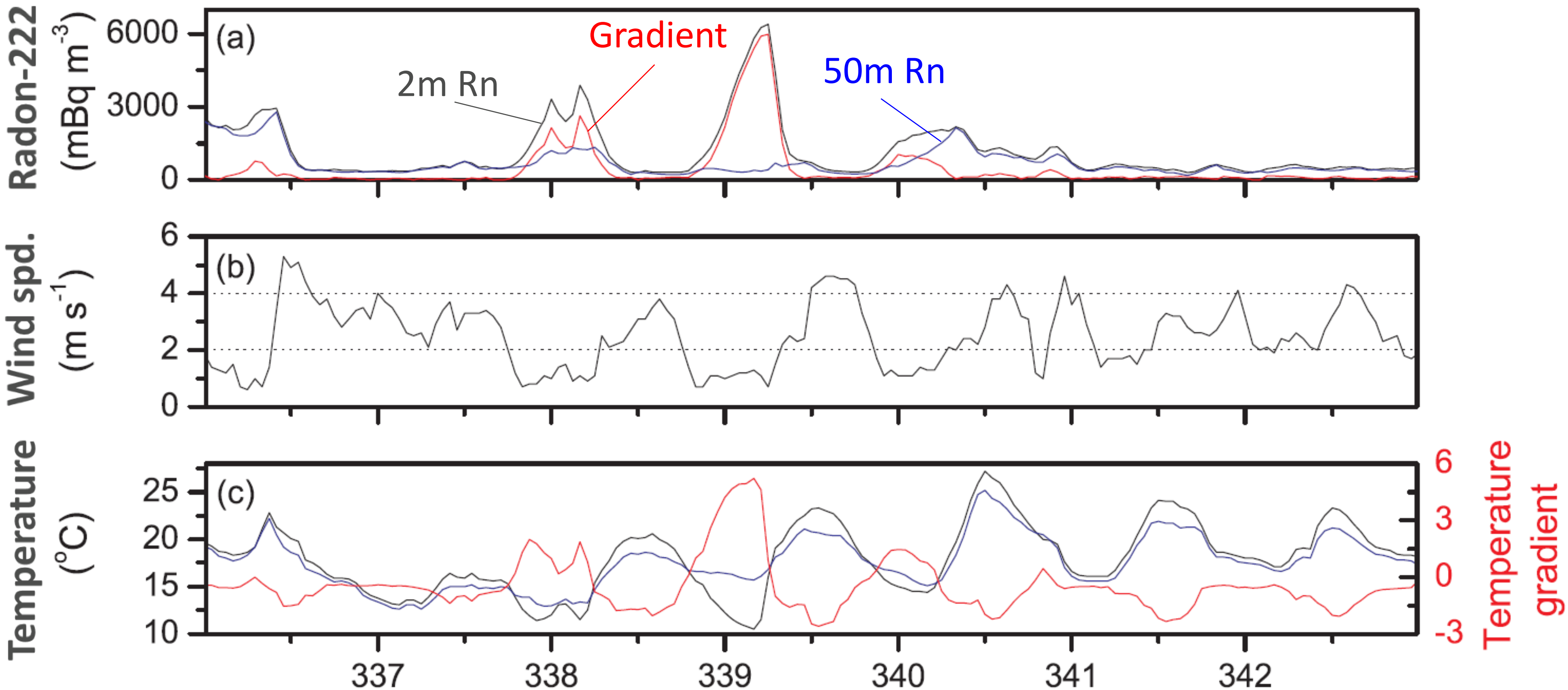
# Simultaneous Rn & meteorological observations




When near surface wind speed drops to ≤1.5 m $s^{-1}$ an accumulation of $^{222}$Rn is evident between 2 and 50 m, as would also be expected of primary pollutants.

Intermittent nocturnal mixing events (e.g. nocturnal jets near the inversion; Williams et al, (2013; DOI 10.1007/s10546-013-9849-3), can interrupt $^{222}$Rn accumulation (e.g. on the morning of day 338).

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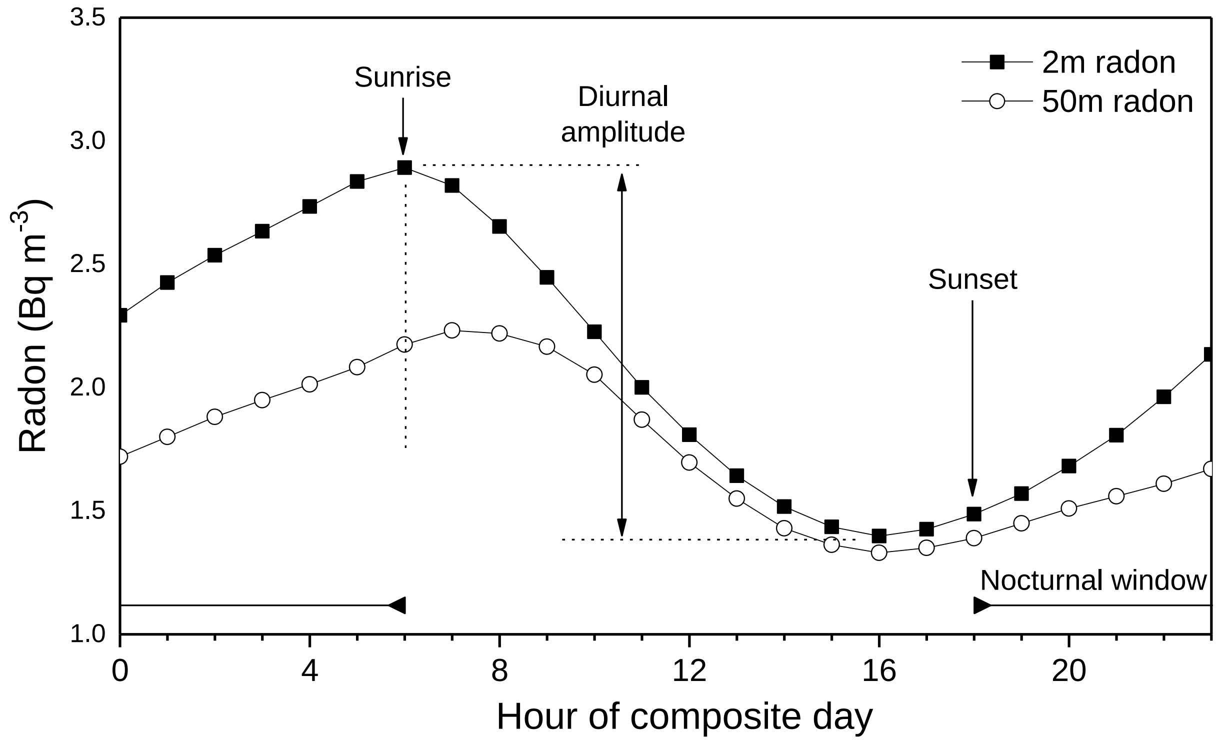
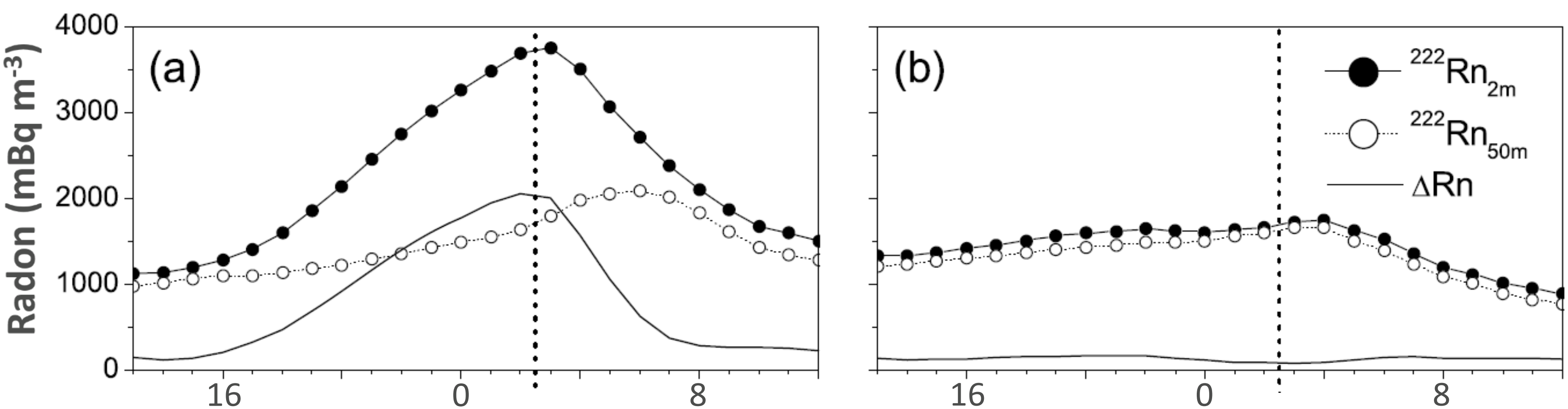
# Near surface vertical $^{222}$Rn gradients

**Right**: example of composite diurnal cycles of $^{222}$Rn at 2 heights (2–50m agl)

**Below**: 2-height Rn observations and the Rn gradient (ΔRn) for (a) a near calm night, and (b) a windy night



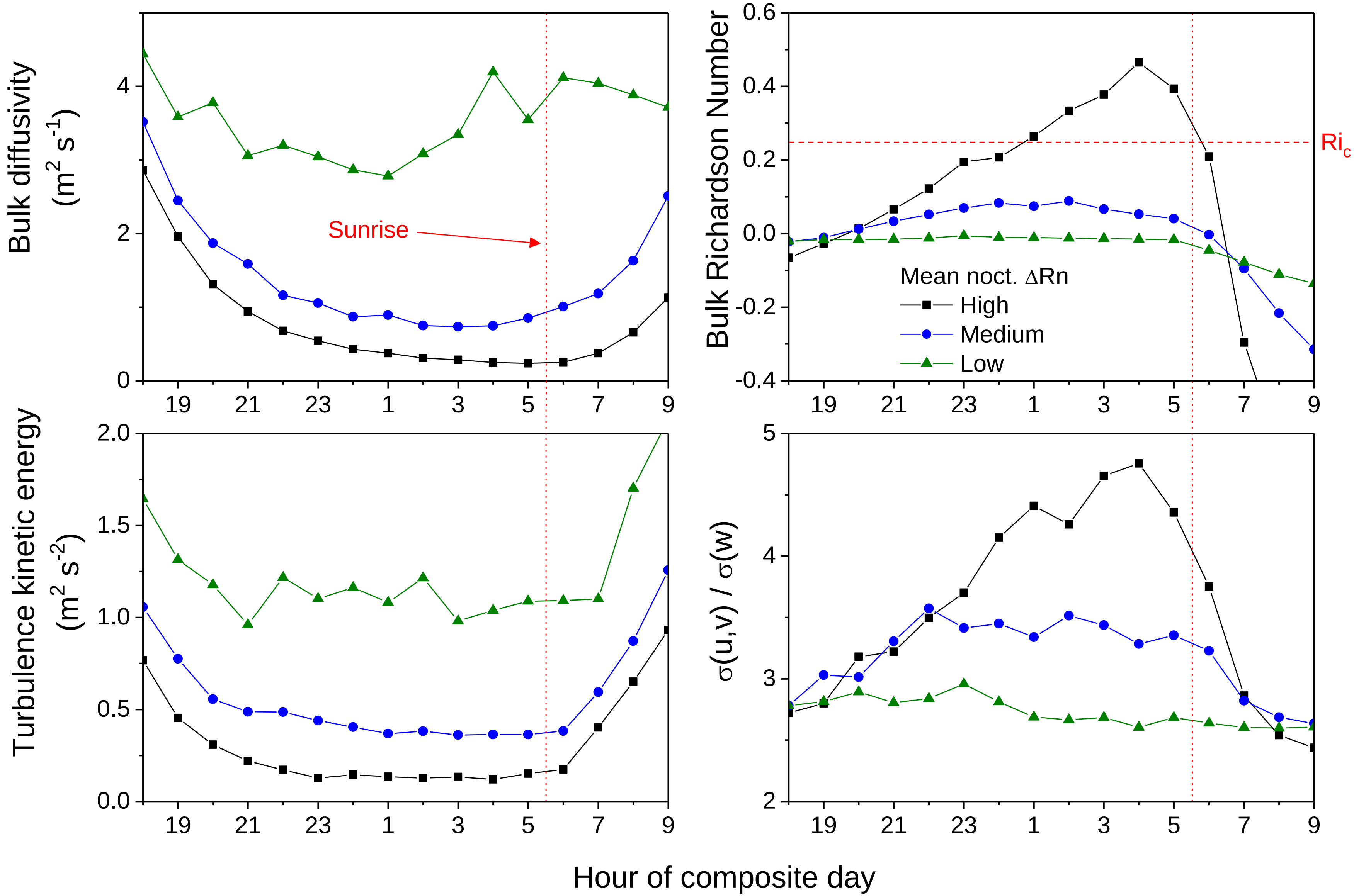



# Notes for previous slide

**Top figure**: Diurnal cycles of radon at both heights are characterised by an afternoon minimum and morning maximum. The morning maximum is delayed for the upper measurement since it takes time for the morning convection to mix radon accumulated near the surface through the 50 m level (Chambers et al. 2011; Tellus, DOI: 10.1111/j.1600-0889.2011.00565.x).

**Bottom figures**: When mean nocturnal wind speeds exceed 1.5 m $s^{-1}$ near surface radon gradients are very small (typically 0.1 - 0.2 Bq $m^{-3}$ over 48 m). The measurement site was on a topographic rise, so nocturnal radon accumulation on calm nights was not very large.

# Relating ΔRn to near surface mixing



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If meteorological quantities (or derived indices of stability) in the stable nocturnal boundary layer are grouped into composites based only on the average radon gradient over the nocturnal window (1900 – 0600h) as shown, they exhibit relatively consistent behaviour within the groups.

Diurnal composites (based on hourly mean values) help to explain the factors that are leading to the near-surface radon accumulation when it occurs.

Nights exhibiting the highest near-surface radon gradients had the lowest turbulent kinetic energy and bulk diffusivity values; likewise, for more than half of these nights the bulk Richardson Number was close to or above the critical value ($Ri_c = 0.25$), indicative of a transition to stability.

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# How essential is ΔRn for mixing studies?

- Clearly, near-surface radon gradients <u>alone</u> (i.e. independent of any other observations) are capable of grouping nocturnal periods whose meteorological mixing conditions are relatively consistent
- However, reliably measuring vertical $^{222}Rn$ gradients across a range of meteorological conditions requires multiple, highly-sensitive, highly-accurate, and therefore relatively expensive, direct (not by-progeny) radon detectors
- 1500L dual-flow-loop two-filter $^{222}Rn$ detectors manufactured by ANSTO have a detection limit of 0.025 Bq $m^{-3}$, with an absolute uncertainty of 10-15% at concentrations around 0.1 Bq $m^{-3}$; but they are roughly 3 times more expensive than an *AlphaGUARD* monitor (detection limit ~2.0 Bq $m^{-3}$)
- If a radon gradient could be *approximated* with a single detector, the slight reduction in accuracy of the classification would be offset by a substantial reduction in cost (and less accurate detectors could be used)
- Making the assumption that observed near-surface radon variability is dominated by diurnal changes in mixing (which is usually the case far from the coast, or when there is no synoptic stagnation of air masses) it is indeed possible to APPROXIMATE a radon gradient measurement in the ABL with a single radon detector

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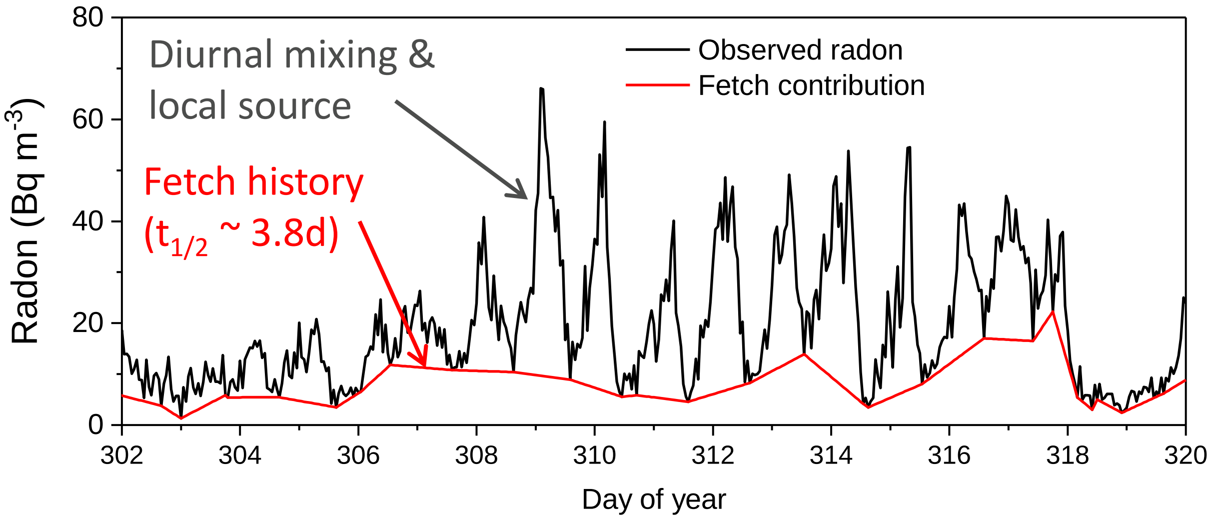
# Approximating ΔRn in the ABL

Imagine two radon detectors, one sampling ~2m agl, the other sampling at a height typically above the nocturnal inversion (e.g. 300-500m)

During the day (when ABL is well mixed), both would report similar values (to within ~0.1 Bq $m^{-3}$), but the upper detector would not "see" the nocturnal radon accumulation (since it would be in the residual layer at night)

In practice, the higher radon observations can be *approximated* by linearly interpolating the 2 m radon observations between afternoon minimum values (this interpolated series is an *approximation* of the radon fetch effects)

The pseudo-gradient, calculated as the difference between the 2 m radon and interpolated afternoon values, can be used for near-surface mixing studies




A radon time series has **mixing** & **fetch** related components

Estimate and remove the fetch component (pm-pm interpolation)

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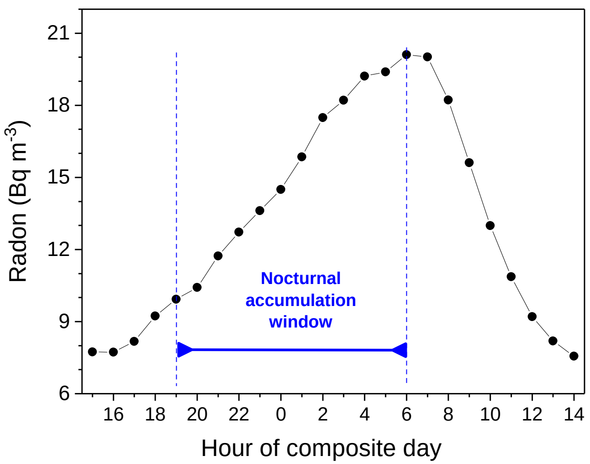
There are three particular conditions under which the necessary assumptions for this radon gradient technique are **not met**:

(i) for measurements within ≤15 km of a coastline (since the large change in surface radon flux can result in observed changes in near-surface radon concentration within the 8 – 10 h nocturnal window that are **not** related to mixing processes);

(ii) during periods of strong synoptic non-stationarity (frontal passages etc.) the calculated nocturnal pseudo-gradient can go largely negative (such periods need to be isolated/removed); and

(iii) during periods of synoptic stagnation (which can persist in some locations and seasons for days to weeks) diurnal mixing may no longer be the dominant driver of observed near-surface variability in radon concentrations.

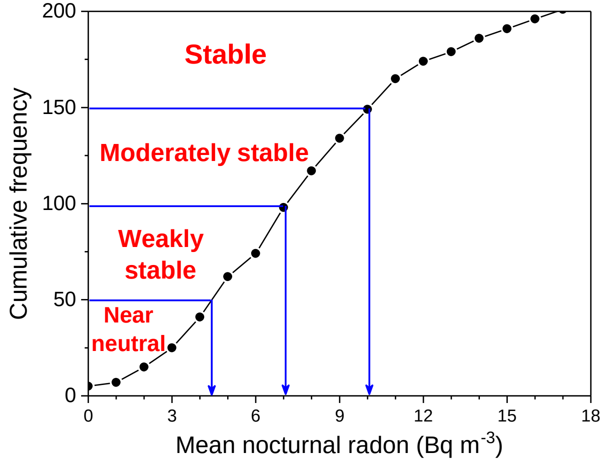
# Pseudo-gradient ($\Delta_P Rn$) as a diurnal stability indicator




Calculate the average $\Delta_P Rn$ each night between 1900 – 0600h

Make a histogram for each season

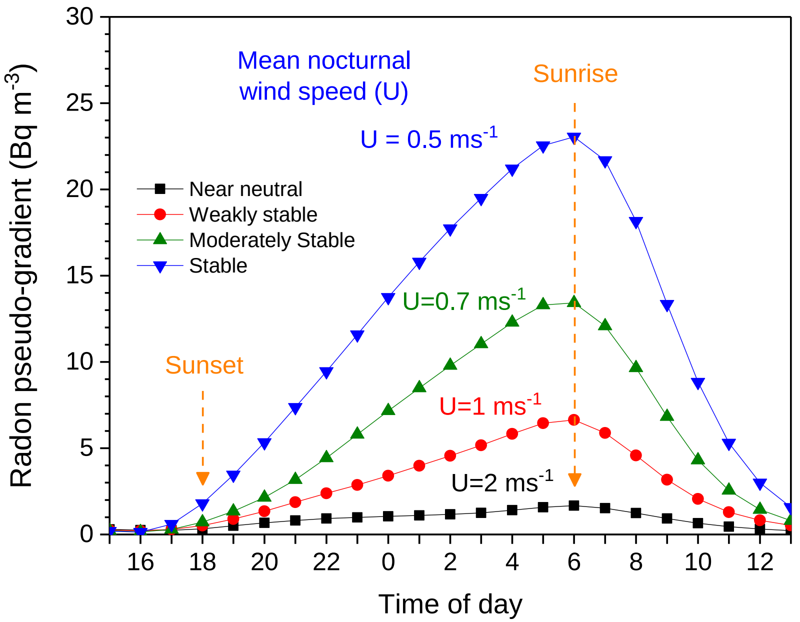
Divide the histogram into nocturnal mixing categories




Assign each mixing category to whole 24h periods. Short-term atmospheric persistence *usually* means conditions that give rise to the observed nocturnal mixing state were similar for the previous evening and following morning

When radon observations are grouped by these categories, diurnal composites (right) show consistent behavioural changes

Meteorology, derived quantities, trace gas or aerosol observations, can all be assigned to these radon-derived mixing categories



# Notes for previous slide

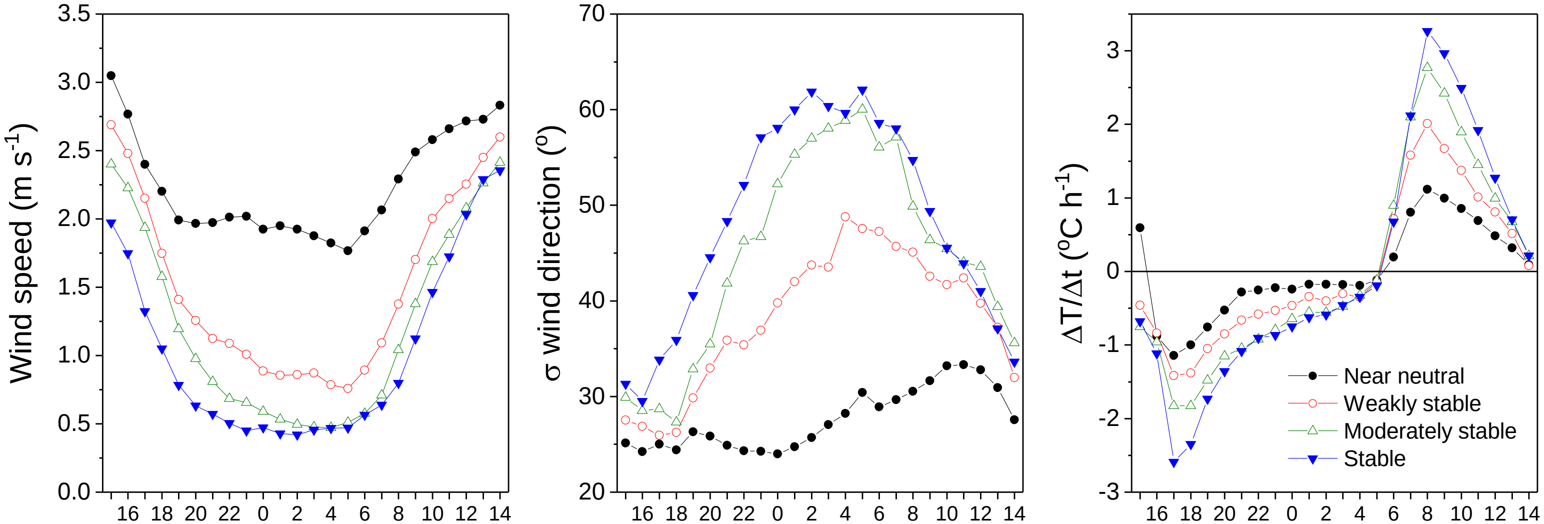
This approach was initially proposed by Chambers et al. (2015; ACP, doi:10.5194/acp-15-1175-2015).

Mixing categories near the surface (in the nocturnal boundary layer) are defined by measurements over an 11-hour nocturnal window.

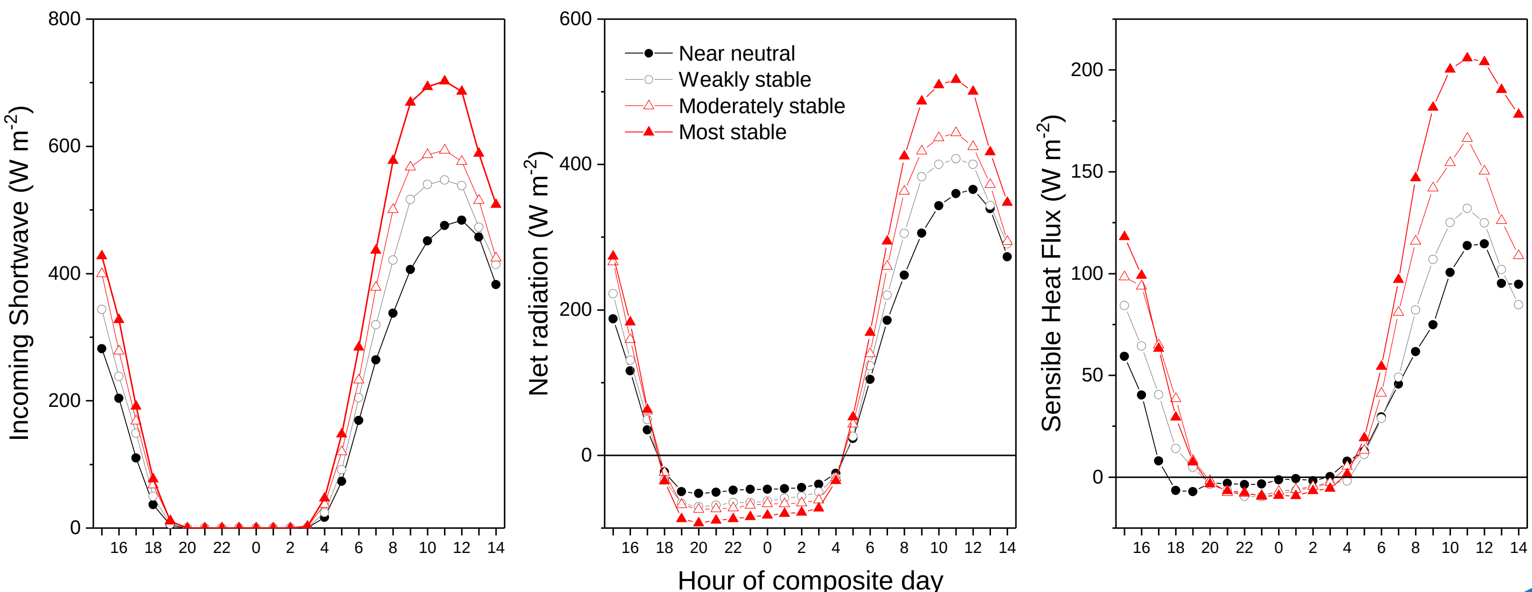
Mixing categories are then applied to whole 24-hour periods, where the mixing conditions of the daytime periods are inferred (not measured) based on an assumption of atmospheric persistence (see Table 1 of Chambers et al. (2019; JGR, doi:10.1029/2018JD029507) for a more detailed explanation).

Assigning mixing categories in this manner implies a night-day crossover of actual atmospheric mixing behaviour, since the conditions giving rise to the most stable nights (anti-cyclonic, low geostrophic winds, clear sky), typically result in the strongest daytime convective mixing.

# Relating $\Delta_P$Rn to site meteorology




NSW, Australia




Europe (Poland)

**Recall**: all classification was performed independently of site meteorology

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# Notes for previous slide

As for vertical two-point radon gradient measurements, classification of the near-surface atmospheric mixing state based on nocturnal radon pseudo-gradient measurements (using only single height radon observations) also proves to be very effective.

In Richmond (NSW, Australia), independently measured stability indicators (including wind speed, standard deviation of wind direction, near-surface temperature gradient (not shown) and rate of change of near-surface temperature), could be consistently grouped using the simplified (1-height) radon-based classification scheme.

To demonstrate the efficacy of the "atmospheric persistence assumption" (which typically holds true ~80% of the time), used to justify applying mixing categories to whole 24-hour periods (each containing 1 whole night), solar & net radiation measurements and sensible heat fluxes from central Poland, classified in the same way, support the prior assertion that conditions responsible for the most stable nights give rise to the most convective days, whereas near-neutral nights are often associated with more overcast days.

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# Timescales of stability influences

- The mixing state of the lower atmosphere changes on a range of timescales

  **Seasonal**: linked to solar elevation, day length & soil freezing
  **Synoptic**: linked to subsidence or uplift in weather systems
  **Diurnal**: linked to the diurnal solar cycle

- The basic **$\Delta_P$Rn** stability classification method addresses <u>diurnal changes</u>. If day length and moisture/freezing influences on the $^{222}$Rn source function are accounted for, seasonal changes <u>can</u> be quantified

- However, substantial synoptic-timescale changes in [$^{222}$Rn] resulting from changes in the atmospheric mixing state violate important assumptions of the basic $\Delta_P$Rn approach

- Such situations arise during synoptic stagnation events, or in continental mid- to high-latitude regions (Europe, and central North America) in the colder months (e.g. mid-autumn, winter to early-spring) when "persistent temperature inversion" conditions can develop
(e.g. in association with the "Siberian High")

# Notes for previous slide

A detailed description of applying the $\Delta_P Rn$ classification method separately by season is provided in Williams et al. (2016; Tellus, http://dx.doi.org/10.3402/tellusb.v68.30967).

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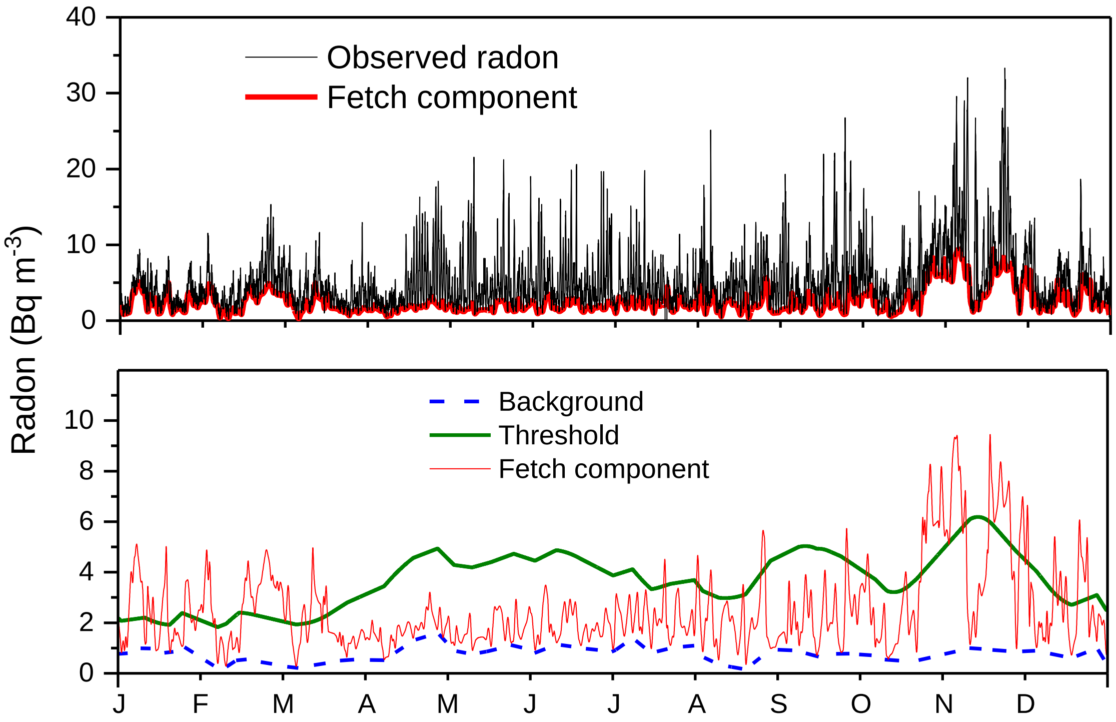
# Identifying synoptic stability changes with $^{222}Rn$

Starting with 1-year of hourly near-surface [$^{222}Rn$] measurements




- Calculate $^{222}Rn$ fetch-effects by interpolation (pm to pm)
- Estimate synoptic timescale for warm/cold months by (spectral analysis of wind or pressure)
- Calculate regional "background" $^{222}Rn$ by interpolating fetch minima on synoptic timescales
- Add monthly σ of diurnal $^{222}Rn$ signal to the background to get the **threshold** value

- When the "fetch component" is > the **threshold** value, synoptic mixing influences on [$^{222}Rn$] exceed diurnal mixing influences
- If this state persists for >48 hours a synoptic timescale "Persistent Temperature Inversion" (PTI) event is occurring

# Notes for previous slide

A detailed description of identifying synoptic timescale "persistent temperature inversion" events using near-surface radon observations is given in Kikaj et al. (2019; AMT, https://doi.org/10.5194/amt-12-4455-2019).

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# Combined Rn-based stability classification

Example application of Rn-based stability classification in Poland based on 4-years obs.

**1.** Identify and isolate PTI events; and **2.** apply the $\Delta_P$Rn method to remaining data

**Table 1: Average number of days per season (composite year) within each atmospheric mixing category. Total number of events per category over the 4 years shown in parentheses.**

| Mix Cat. | Comment | Winter | Spring | Summer | Autumn |
|---|---|---|---|---|---|
| 1 | Moderate to strong or gusty winds, **non-stationary** synoptic conditions (fronts, etc.), rainfall common **(excluded)** | **12** (48) | **17** (68) | **16** (64) | **15** (60) |
| 2 | Moderate to strong winds, often overcast, rainfall common **(near-neutral)** | **13** (52) | **17** (68) | **16** (64) | **14** (56) |
| 3 | Light to moderate winds, cloudy to overcast, occasional rainfall **(weakly stable)** | **12** (48) | **17** (68) | **15** (60) | **14** (56) |
| 4 | Light winds, scattered cloud (mostly clear), rainfall uncommon **(moderately stable)** | **13** (52) | **17** (68) | **16** (64) | **15** (60) |
| 5 | Calm to light winds, mostly clear skies, convective daytime rain if any **(most stable)** | **13** (52) | **18** (72) | **16** (64) | **15** (60) |
| 6 | Near-calm, fog/haze common **(PTI)** | **27** (108) | **5** (20) | **0** (0) | **18** (72) |

- 5 diurnal categories defined for this study (15-20% of each season)
- 1 synoptic category: PTI events 5-30% of non-summer months

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Detailed explanations of this combined timescale approach to radon-based classification of the atmospheric mixing state are given in:
Chambers and Podstawczynska (2019; Atmos. Environ, https://doi.org/10.1016/j.atmosenv.2019.117040)
and
Kikaj et al. (2019; Atmos. Res., https://doi.org/10.1016/j.atmosres.2019.104838).

It should be stressed, however, that in coastal regions prone to atmospheric stagnation-related poor air quality events (e.g. eastern Po Valley region) this approach is not applicable (due to the strong spatial heterogeneity of the radon source function).

# Benefits of Rn-based classification of the atmospheric mixing state

- Urban climate and urban air quality are both influenced by a combination of "controllable" and "uncontrollable" influences
- **Controllable** influences include source strengths, types & their spatial distribution; urban design (energy efficiency, ventilation, filtration, etc.)
- **Uncontrollable** influences include meteorology; topographic setting; pollution advection from other regions (etc.)
- To develop and assess urban climate and air quality mitigation measures it is essential to be able to disentangle these effects (since, by definition, mitigation can only target controllable influences)
- Atmospheric "class-typing" using $^{222}Rn$ facilitates this separation
- By grouping periods of more consistent/comparable mixing conditions, controllable influences can be the subject of more targeted investigation
- The robust statics afforded by seasonal application of Rn-based classification accounts for spatial and temporal source heterogeneity, helping to bridge the scale gap between observations & simulations for evaluation of regional model performance

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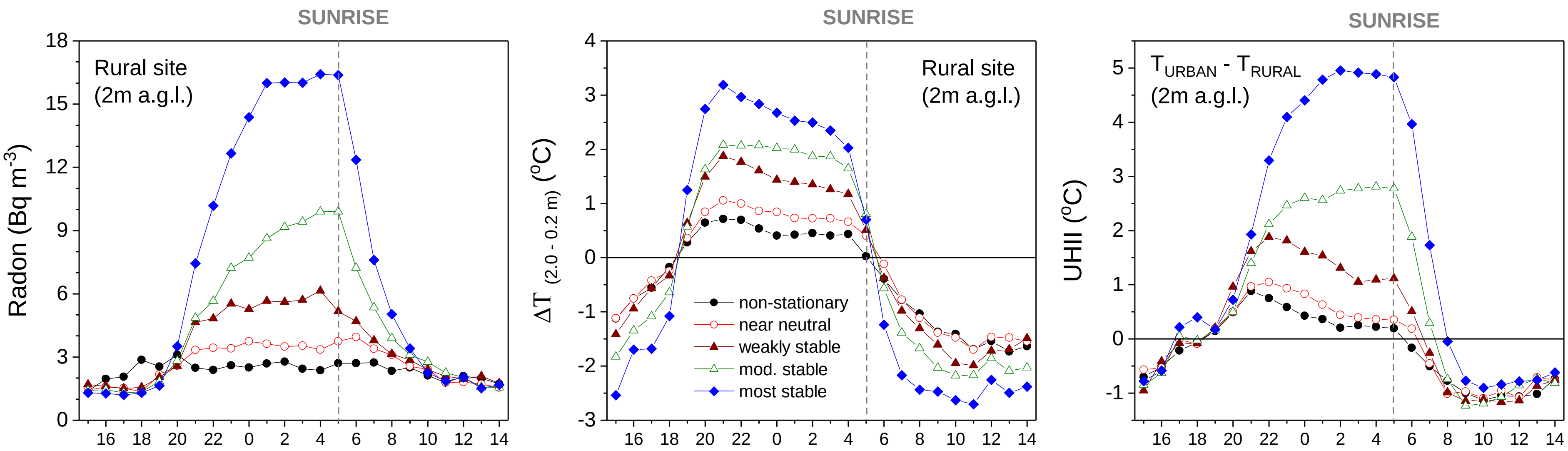
# Notes for previous slide

Examples of ways in which Rn-based atmospheric class typing can assist with the evaluation of regional chemical transport model performance are given by Chambers et al. (2019; Atmosphere, doi:10.3390/atmos10010025).

# Rn-based class typing for urban climate studies




- In Lodz (central Poland), $^{222}$Rn was used to classify the atmospheric mixing state in a background rural setting 25 km from the city centre
- Testimony to the fidelity with which the radon class types group like stability states, composites of the near surface temperature gradient (between 0.2 and 2m) demonstrated consistent variation between classes
- Likewise, the observed **Urban Heat Island Intensity** (defined as the 2m temperature difference between the city centre and background site) exhibited distinct behaviour between, and consistent behaviour within, the defined mixing states

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Where multi-year datasets exist, there are sufficient days assigned to each of the stability categories to enable seasonal investigations of other factors contributing to the urban heating effect ***within*** each of the specific mixing categories (thereby eliminating, or greatly reducing, the influence of uncontrollable factors).

More detailed discussion of these points is provided in Chambers et al. (2019; JGR, https://doi.org/10.1029/2018JD029507).

These results indicate that radon-based atmospheric class typing from a **single well-chosen site**, can be representative of a broad region of relatively simple topography, and be used to simultaneously interpret measurements from a broad network of urban climate sensors.

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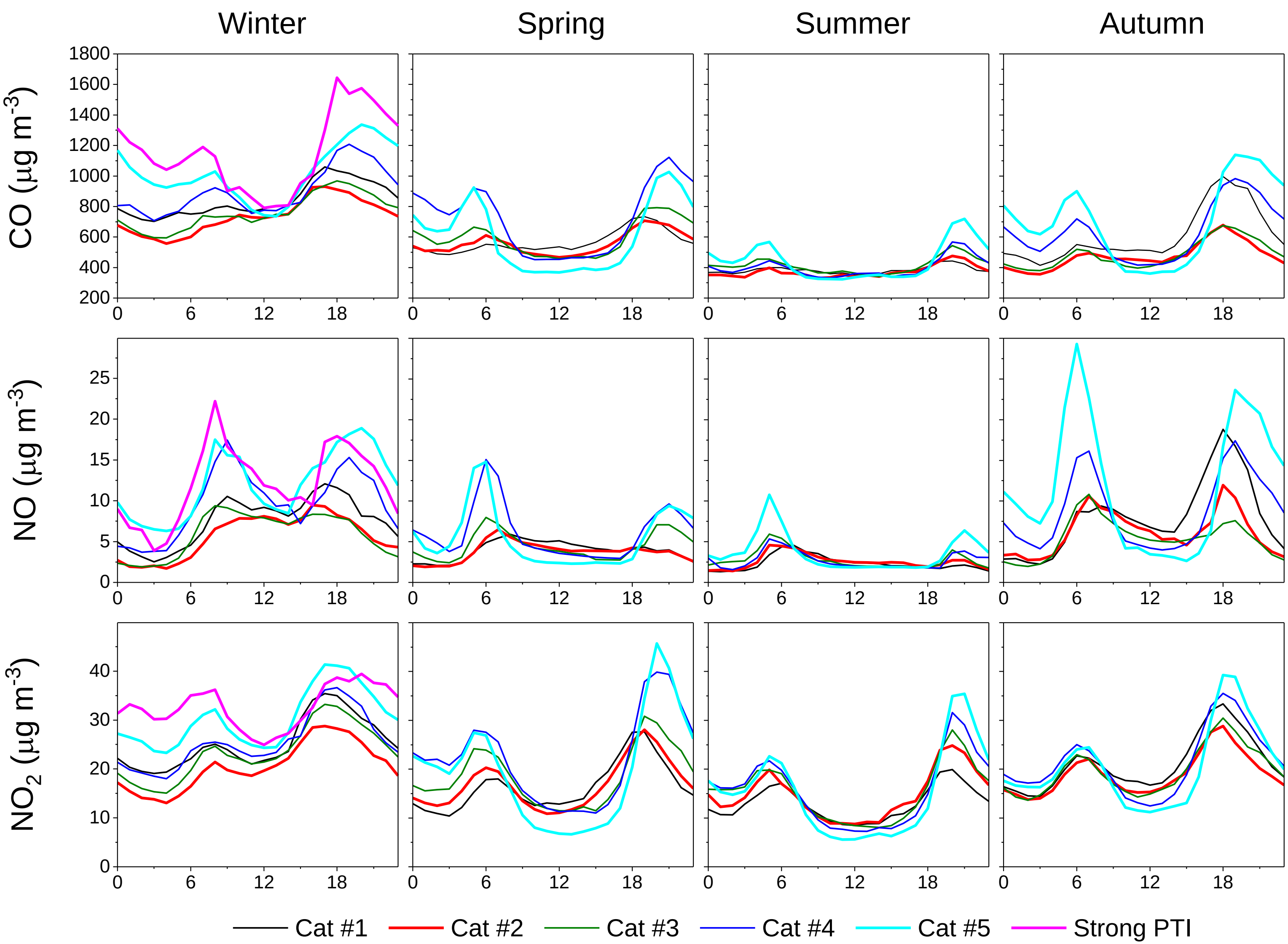
# Rn-based class typing for air quality studies




Statistically robust information about the influence of particular mixing conditions on the variability of traffic emissions can be derived quite simply for all seasons.

Comparing characteristics of the more consistent mixing conditions (eg. Cat 4,5) from year to year, can help gauge the efficacy of mitigation measures.

**Traffic pollution in Zgierz, a northern suburb of Łódź (Poland)**

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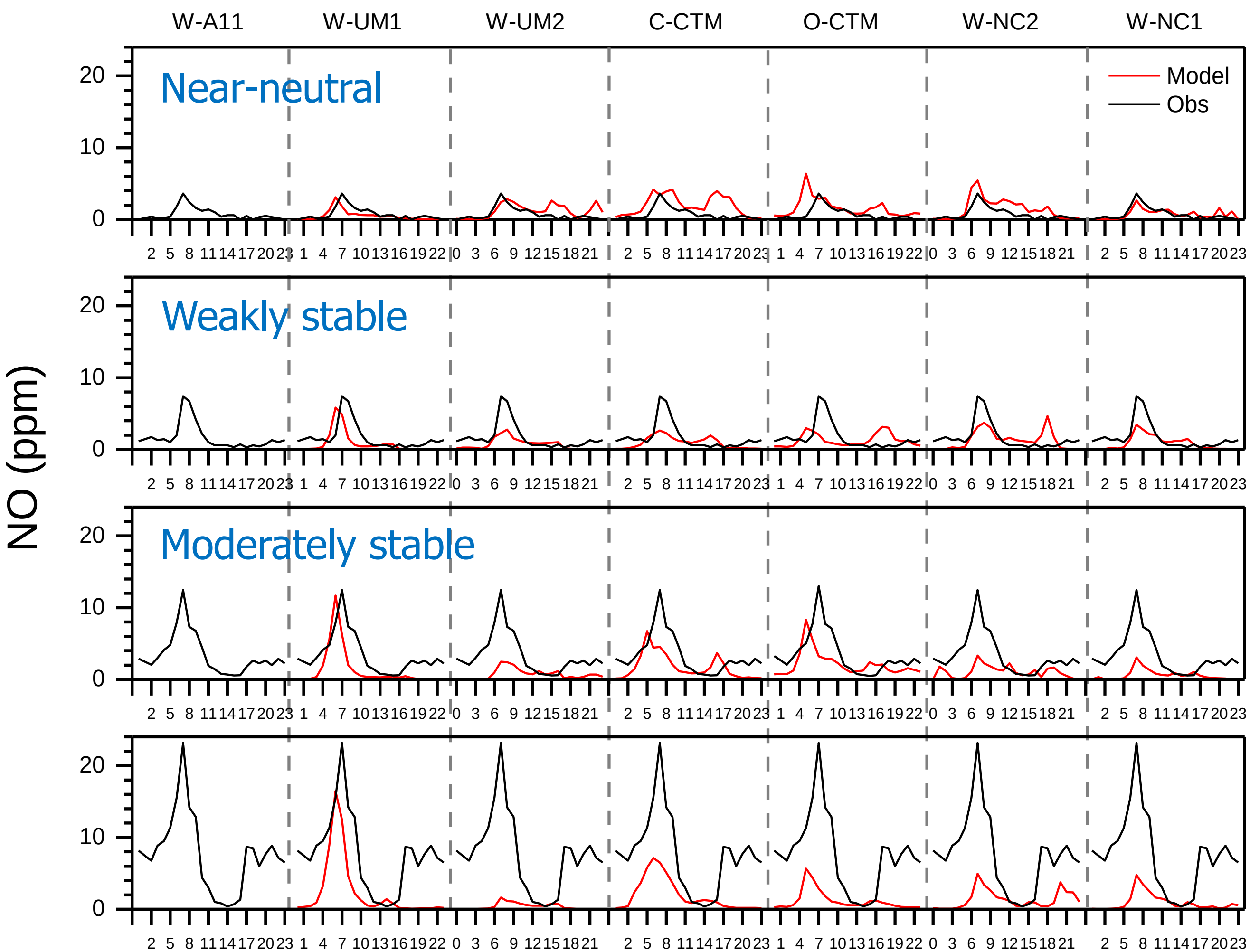
The typically strong seasonality in traffic emission concentrations in urban regions can more readily be related to changes in mixing depths with the assistance of Rn-based class typing (see Williams et al 2016; Tellus, http://dx.doi.org/10.3402/tellusb.v68.30967).

Furthermore, while the more stable mixing categories (Class 4, 5 and PTI), can be used to perform top-down emission estimates, assess the need for or test the efficacy of mitigation measures, and help assess public health risks, the well-mixed conditions (Classes 1 – 3) can be used to help assess pollution advection from remote or neighbouring regions.

For further information see Podstawczynska and Chambers (2019; Atmos. Environ., https://doi.org/10.1016/j.atmosenv.2019.117038).

# Skill testing CTMs: primary pollutant example

- Separated observed and simulated NO by mixing class (Sydney, Aust)
- Model performance was best when atmosphere was well mixed




Most Stable

Sensitivity of model skill to mixing state was different between models (all using similar emissions inventories)

Most models performed worst under stable conditions (when public exposure was highest)

# Notes for previous slide

Evaluating the performance of 7 chemical transport models for vehicle emissions in the Sydney Basin, Australia.

The models tested were:
**W-A11** (ANSTO, WRF-Chem v3.7.1, with radon),
**W-UM1** (UoM, CMAQ with WRF v3.6.1, with radon),
**W-UM2** (UoM, WRF-Chem v3.7.1),
**C-CTM** (CSIRO, CCAM & CTM - Australia's AQ Forecasting Model),
**O-CTM** (NSW OEH, "Operational" CCAM and CTM),
**W-NC1** (North Carolina State Uni, WRF-Chem v3.7.1), and
**W-NC2** (NCSU, WRF-Chem v3.7.1 coupled to ROMS).

For further information see:
Chambers et al (2019; Atmosphere, doi:10.3390/atmos10010025),
Guérette et al. (2020; Atmosphere, DOI: 10.3390/atmos11030233), and
Monk et al., (2019; Atmosphere, doi:10.3390/atmos10070374).

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# Rn-based class typing: scales of applicability

- The radon pseudo-gradients used here to perform seasonal atmospheric mixing "class-typing" on diurnal and synoptic timescales have all been based on near-surface observations from a single location; but how applicable are they beyond the local stable nocturnal boundary layer?
- To better assess the utility of this technique in terms of its suitability to simultaneously help to interpret climatic or air quality data from a broad network of sensors (throughout the urban canopy layer), or evaluate a chemical transport model throughout the urban boundary layer, it is necessary to assess the scales of applicability (horizontally and vertically)
- This has been attempted in two ways:

    1. In central Poland we formed composite 6am synoptic mean sea level pressure charts according to the radon-based mixing categories, and

    2. In central Slovenia we formed composite 6am radio-sonde profiles according to the radon-based mixing categories

- Both spatially and vertically the radon-based technique grouped conditions of comparable characteristics in a consistent way

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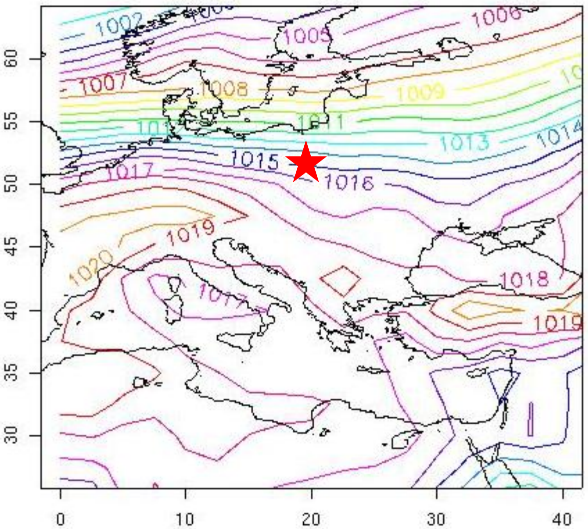
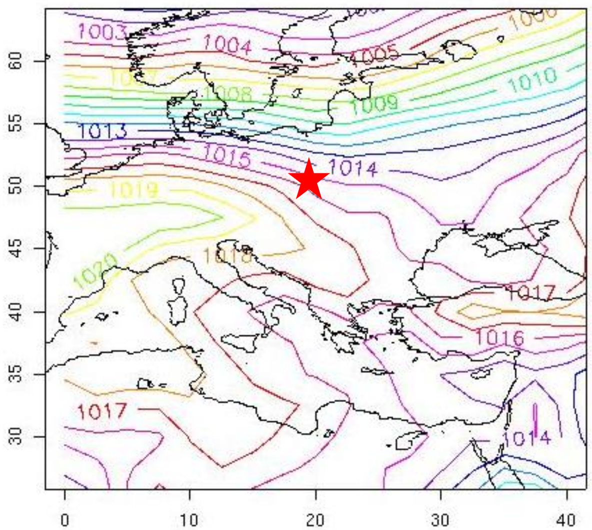
# Synoptic setting for Rn mixing classes

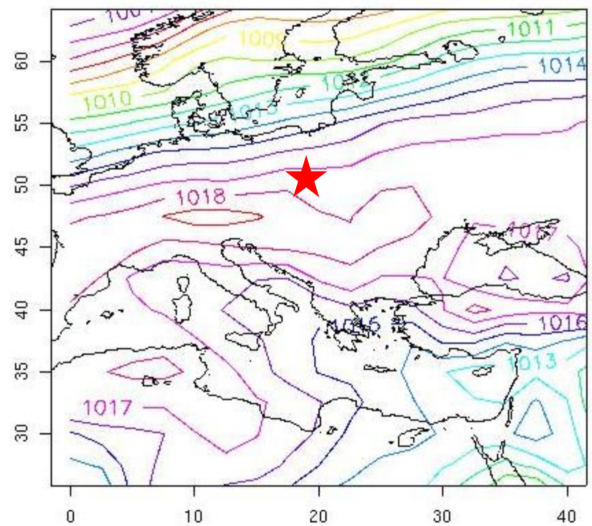
**Class #2: Well-mixed nights**



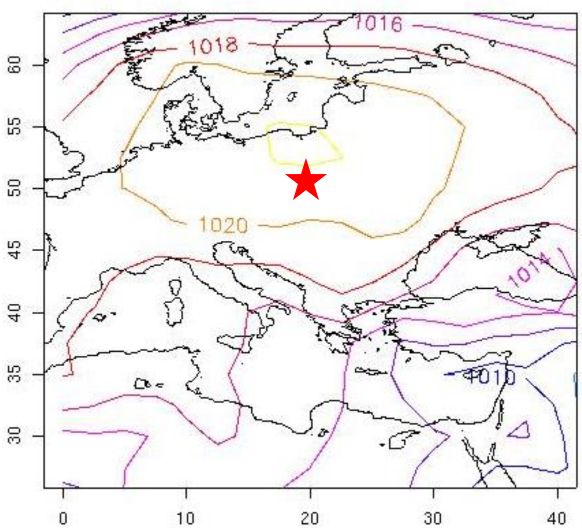
**Class #3: Weakly stable nights**



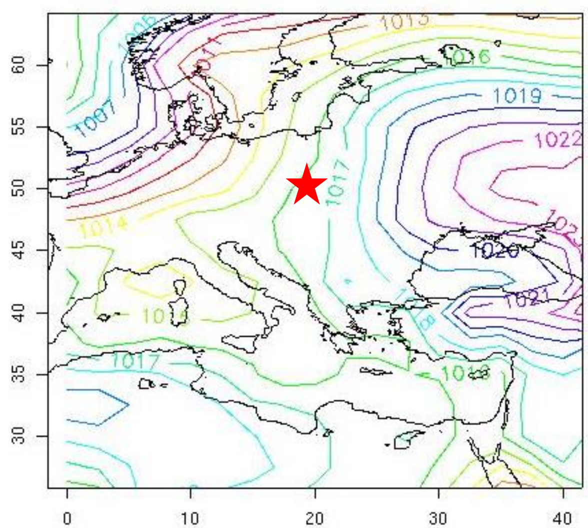
**Class #4: Moderately stable**



**Class #5: Most stable nights**



**Class #6: PTI events**



PTI events occur when the **Siberian High** is well-established.

Regional scale flow is slow and from the south (across Europe) accumulating primary emissions.

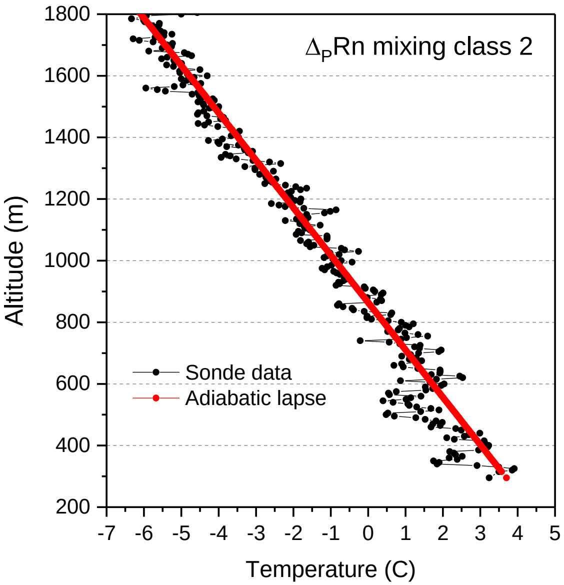
# Notes for previous slide

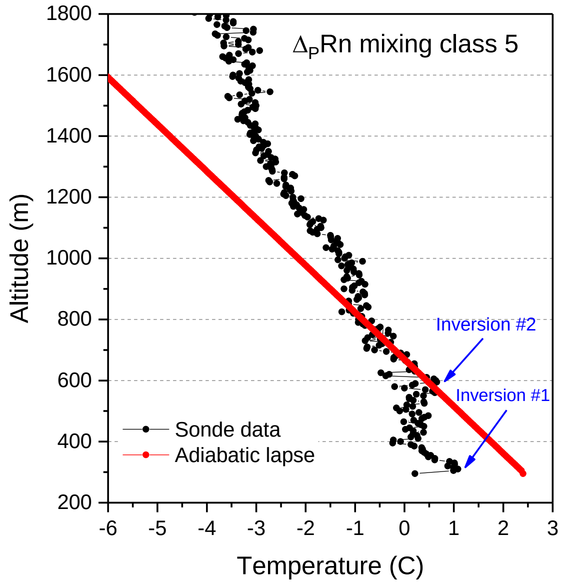
For observations in central Poland, the most well-mixed and overcast / inclement conditions were associated strong north-westerly winds coming to the site fairly directly from the North Sea.

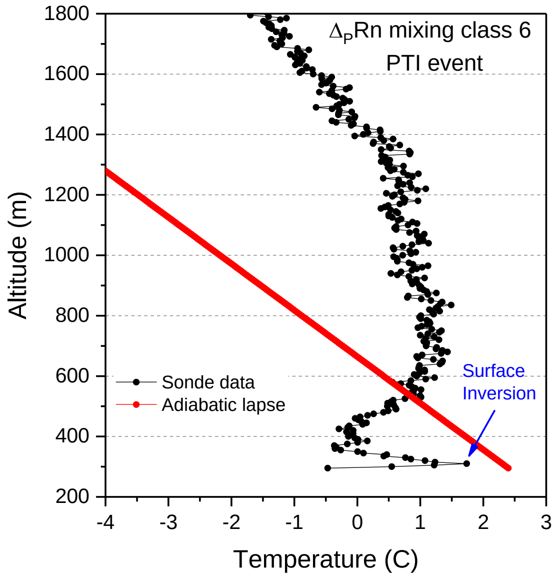
On diurnal timescales, the most stable nights were associated with an anti-cyclone situated directly over the site.

On synoptic timescales, the most stable (and most polluted) conditions were associated with slow southerly flow driven by a well-established “Siberian High” to the east.

# Rn-based class typing: vertical extent




- A composite of all 6am sonde profiles from Ljubljana (Slovenia) under near-neutral (class 2) conditions yielded a temperature profile close to adiabatic
- A composite of 6am profiles under class 5 conditions (local anti-cyclone influence) shows a surface inversion, and second inversion near the height of the lowest mountain peaks of the Ljubljana basin
- A composite of 6am profiles under PTI conditions shows very stable air within the basin, and the atmosphere tending toward stability for another 1 km above the basin

# Conclusions

- Near-surface radon observations are closely linked to the net effect of vertical mixing processes near the surface
- Continuous outdoor radon measurements can be used for atmospheric class-typing at the urban scale on diurnal and synoptic timescales
- Rn-based atmospheric class-typing provides statistically robust datasets for the whole diurnal cycle with reduced variability
- Useful for health studies, developing and testing mitigation measures, and evaluating pollution models for urban planning
- Simple, consistent, universal and economical technique

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