## 9B.2 DIURNAL BOUNDARY LAYER MIXING PATTERNS CHARACTERISED BY RADON-222 GRADIENT OBSERVATIONS AT CABAUW

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# **1. INTRODUCTION**

Accurate representation of the atmospheric boundary layer, and the nocturnal boundary layer in particular, through which most energy, trace gas, and aerosol exchanges take place, continues to be problematic in contemporary weather and climate models. Until this issue has been adequately addressed our ability to forecast future climate trends with reasonable confidence will be compromised by the large differences observed between measured and modeled near surface climate parameters, and indeed, even between those of comparable models – particularly in high latitude regions.

To evaluate, and ultimately improve, boundary layer mixing schemes in such models it is first necessary to construct quantitative measures of vertical mixing and exchange within the lower atmosphere at a temporal resolution sufficient to resolve the diurnal cycle. One way to quantitatively characterize near-surface mixing processes on diurnal time scales is to make continuous, high temporal resolution, gradient measurements of a suitable atmospheric tracer.

Radon is a naturally occurring, radioactive, noble gas, with a low solubility in water. It is emitted from terrestrial surfaces at an approximately constant rate on diurnal timescales and the emissions are assumed to be uniform on local to regional scales (Jacob et al., 1997). The half-life of radon (3.8-days) is optimum for boundary layer mixing studies, since it is long compared with typical turbulent timescales ( $\leq$ 1-hour) but short enough to constrain its concentration in the free troposphere to be typically 1-3 orders of magnitude lower than near surface values. The combination of these properties makes radon an excellent tracer for vertical mixing studies in the boundary layer.

## 2. METHODS AND SITES

In this study we measured atmospheric radon concentrations using 1500 L dual flow loop, two filter radon detectors (Zahorowski et al., 2005). This kind of detector is designed for multi-year deployment, provides continuous hourly radon concentration data, and has a lower limit of detectors can not be multiplexed for gradient measurements, due to their 45-minute response

time, so dedicated detectors are used for each height of the gradient measurements.

At present we are measuring near-surface radon gradients from meteorological towers at Lucas Heights, New South Wales (50 m tall; 150.981°E) 34.053°S, and the Cabauw Experimental Site for Atmospheric Research (CESAR) in The Netherlands (213 m tall; 51.971°N, 4.927°E). The Lucas Heights gradient observations, between 2-50 m above ground level (agl), specifically target mixing in the atmospheric surface layer whereas those at CESAR, between 20-200 m agl, target mixing in the lower atmospheric boundary layer.

This discussion focuses on observations at CESAR only, which is located 50 km inland on a polder in an agricultural region. The site is topographically simple, with surface elevations changing by only a few metres within a 20 km radius. Preliminary results from the first year (2007) of radon gradient observations at CESAR are presented, with an emphasis on diurnal, synoptic and seasonal timescales. Supporting meteorological data for this period was provided by the Royal Netherlands Meteorological Institute (KNMI).

## 3. RESULTS AND DISCUSSION

#### 3.1 Seasonal Variability

A pronounced variability in the seasonal median radon concentration was evident at both sampling heights (Figure 1). The lowest concentrations were observed in winter and summer, when the dominant air mass fetch was the Atlantic Ocean, either via the English Channel, or across the British Isles. The highest radon concentrations were observed in spring and autumn, when a larger proportion of air mass fetch was over western and/or central Europe.

Based on wind direction frequency analyses, approximately 40% of autumn air masses arriving at CESAR had recent mainland European fetch compared to only 26% in summer. Furthermore, when air mass fetch was oceanic in spring and autumn, it was often over the North Sea, leeward of the British Isles, where radon concentrations are likely to be perturbed from usual oceanic baseline values ( $\leq 100 \text{ mBg m}^{-3}$ ).

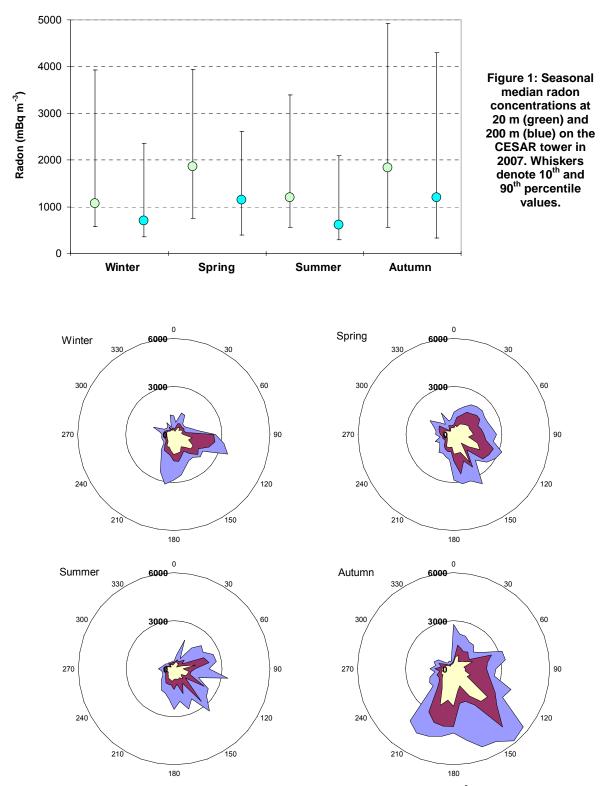


Figure 2: Seasonal angular distribution of 200 m radon concentration (mBq m<sup>-3</sup>) at CESAR; including lower quartile values (cream), median (magenta) and upper quartile (blue).

The mainland European fetch, approximately represented by wind directions between 50° and 220°, had a pronounced impact on observations at CESAR (Figure 2). For example, in autumn, median radon concentrations in the mainland European fetch were 3.3 times larger than corresponding radon concentrations from the Atlantic/North Sea fetch region. In autumn, the 90<sup>th</sup> percentile radon concentration in the mainland fetch (i.e. representative of the most consistent terrestrial fetch) was 5360 mBq m<sup>-3</sup>. By contrast, the 10<sup>th</sup> percentile radon concentration in the oceanic sector in summer (i.e. the most consistent oceanic fetch) was 255 mBq m<sup>-3</sup>.

#### 3.2 Diurnal Variability

A pronounced diurnal variability was also evident in the CESAR radon observations (Figure 3). The diurnal 20 m radon signal, as derived from all observations, was characterized by an early morning maximum, due to the accumulation of radon in the stable nocturnal boundary layer, and an early afternoon minimum, at the time of maximum atmospheric boundary layer depth. Typical amplitudes of the 20 m radon diurnal cycle ranged from 470 mBq m<sup>-3</sup> in winter to 1420 mBq m<sup>-3</sup>

in spring. The only feature of the 200 m diurnal radon signal (not shown) was a mid-morning maximum, evidence of the near surface nocturnally accumulated radon mixing upwards with the morning development of the convective boundary layer.

The observed variability at 20 m was contributed to by two independent signals, which will henceforth be referred to as remote and local. The remote radon signal represents the accumulated influence from all distant radon sources in the air mass' recent (<2-week) history, due to the 3.8-day half life of radon. The local radon signal represents a combination of local radon sources and the concentration/dilution effects of diurnal changes to the local atmospheric mixing depth.

Under low wind speed conditions, typical of summer in the region, local contributions dominate the diurnal radon signal. We separated contributions to the 20 m diurnal radon signal based on arbitrarily defined wind speed thresholds: low <3 ms<sup>-1</sup> and high >7 ms<sup>-1</sup> (Figure 3).

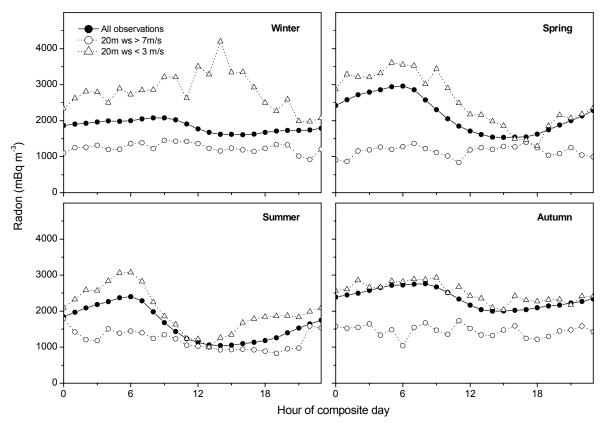


Figure 3: Seasonal diurnal-composite 20 m radon concentrations at CESAR. Filled circles represent all observations, open triangles correspond to low wind speeds (<3 ms<sup>-1</sup>) and open circles correspond to high wind speeds (>7 ms<sup>-1</sup>)

The low wind speed signal exhibited a pronounced diurnal cycle with an average annual amplitude of 1140 mBq m<sup>-3</sup>. At such times nocturnal radon gradients of 2000-5000 mBq m<sup>-3</sup> were commonly observed between 20 and 200 m agl (Figure 4a), due to suppression of turbulent mixing by the nocturnal temperature stratification.

In contrast, the high wind speed signal showed little diurnal cycle, with an annual average amplitude of only 278 mBq m<sup>-3</sup> (Figure 3). Under high wind speed conditions, the nocturnal boundary layer is weakly stratified or absent, and radon is mixed through a deep column leading to a weak diurnal signal (remote contributions dominate).

#### 3.3 Synoptic Variability

During periods of moderate to high wind speed, typical of winter in this region, remote contributions dominated the CESAR radon observations. Mechanical turbulence maintained a substantial mixed layer throughout the nights and only small (0 - 500 mBq  $m^{-3}$ ) gradients were observed between the 20 and 200 m observations (Figure 4b). Under these conditions the primary source of variability in the observed radon signal was due to synoptically driven changes in the recent air mass fetch.

Figure 4b shows an example of a 2-day shift in recent fetch from predominantly Atlantic Ocean, to western Europe. This change results in an enhancement of the observed radon concentration that was comparable in magnitude to the largest of the near-surface nocturnal values in summer.

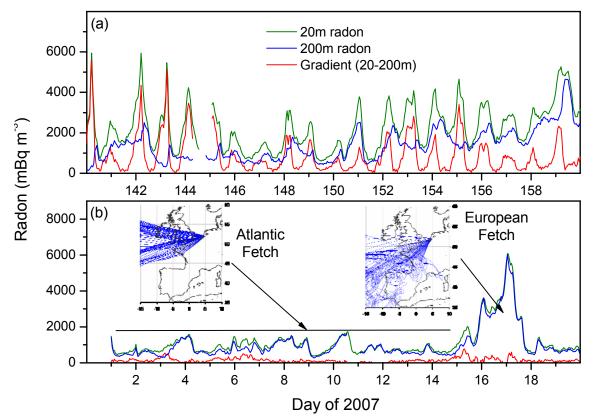


Figure 4: Comparison of radon observations between 20 and 200 m on the CESAR tower (a) when diurnal influences prevail under low wind speed conditions, and (b) when synoptic fetch conditions prevail under high wind speed conditions.

## 4. Conclusions

Our preliminary analysis of the first year of radon observations at CESAR has illustrated the influence on the radon signal of atmospheric processes occurring on three distinct spatiotemporal scales: seasonal, synoptic and diurnal. The 20 m – 200 m radon gradient proved to be a highly sensitive direct measure of the degree of vertical turbulent mixing in the lower atmosphere. Under low wind speed conditions the suppression of turbulence by nocturnal temperature stratification greatly enhances near-surface radon concentrations. In the morning, when the inversion

is burned off, the near-surface radon is diluted in the growing convective boundary layer.

We demonstrated that CESAR radon observations are a combination of local and remote influences, and that in general, by comparing observed radon concentrations between 20 m and 200 m under certain climatic conditions, it will be possible to disentangle these influences on synoptic and diurnal time scales – an important step towards unambiguously characterising and quantify diurnal vertical mixing.

Once estimates of the regional terrestrial radon source function have been made, the next step will be to compare the observed radon time series and gradient observations with a column or regional model under conditions dominated by local and remote sources to evaluate mixing and transport schemes.

#### 5. References

Jacob, D.J., M.J. Prather, P.J. Rasch, R.-L. Shia, Y.J. Balkanski, S.R. Beagley, D.J. Bergmann, W.T. Blackshear, M. Brown, M. Chiba, M.P. Chipperfield, J. de Grandpré, J.E. Dignon, J. Feichter, C. Genthon, W.L. Grose, P.S. Kasibhatla, I. Köhler, M.A. Kritz, K. Law, J.E. Penner, M. Ramonet, C.E. Reeves, D.A. Rotman, D.Z. Stockwell, P.F.J. van Velthoven, G. Verver, O. Wild, H. Yang, and P. Zimmermann, Evaluation and intercomparison of global atmospheric transport models using 222Rn and other short lived tracers, J. Geophys. Res., 102(D5), 5953-5970, 1997.

Zahorowski, W., S. Chambers, T. Wang, C.-H. Kang, I. Uno, S. Poon, S.-N. Oh, S. Werczynski, J. Kim and A. Henderson-Sellers. Radon-222 in boundary layer and free tropospheric continental outflow events at three ACE-Asia sites. Tellus, 57B, 124-140, 2005.

### 6. Acknowledgments

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