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MIXING AND VENTING IN CLEAR AND CLOUDY BOUNDARY LAYERS USING AIRBORNE RADON MEASUREMENTS

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1. INTRODUCTION

Parameterisation of mixing and entrainment processes in the atmospheric boundary layer under clear and cloudy conditions continues to be a topic of intensive research. The accuracy of representations of these processes in regional and global weather and climate models remains a central issue limiting their performance in predicting the distribution of thermodynamic variables and pollutants in the lower atmosphere on diurnal, seasonal and inter-annual timescales. Progress in this field is hampered by a lack of naturally ubiquitous tracer species that are conveniently measurable and have simple and accurately known source and sink functions as well as a spatial distribution that is suitable for unambiguous interpretation of vertical mixing and exchange processes within the lower atmosphere.

Radon-222 is a naturally occurring, radioactive, noble gas, with a low solubility in water. It is produced through the α-decay of Radium-226 and is the only gaseous decay product of the Uranium-238 series. As Radium-226 is ubiquitous to most soil and rock types, this results in a fairly consistent flux of radon from all terrestrial surfaces of between 0.8 – 1.2 atoms cm^{-2} s^{-1}. For the purposes of boundary layer measurements, the radon surface source function can thus be assumed to be approximately constant on diurnal timescales, and the emissions from land surfaces can furthermore be assumed to be horizontally uniform on local to regional scales (Jacob et al., 1997).

As radon is chemically inert, its only significant atmospheric sink is via radioactive decay. The half-life of radon (3.8-days) is optimum for boundary layer mixing studies, since it is long compared with typical turbulent timescales (≤1-hour) but short enough to constrain its concentration in the free troposphere to be typically 1-3 orders of magnitude lower than its near-surface values.

This combination of properties makes radon an excellent tracer for vertical mixing studies in the boundary layer. In the current paper, we present results from field campaigns using light aircraft that demonstrate the use of vertical radon profiles as a quantitative indicator of the degree of vertical mixing and venting under clear and cloudy conditions in the atmospheric boundary layer.

2. METHODS AND MEASUREMENTS

2.1 Airborne radon sampling system

Over the past 4 years, ANSTO has been developing sampling systems for measurement of atmospheric radon concentrations using airborne platforms.

Technical limitations preclude accurate real-time direct measurements of radon in air from light aircraft, because radon's ultra-low atmospheric concentrations dictate a measurement approach using radioactive scintillation counting that requires large delay volumes, resulting in slow response times. It was therefore decided to use a sampling approach utilising charcoal traps to capture radon in-flight, and then to transport the exposed traps back to the laboratory for extraction and counting.

Figure 1: 6-trap airborne radon sampler

The airborne radon sampler used in the current study (Fig. 1) accommodates six (6) charcoal traps connected to a 24V pump via electronically-controlled solenoid valves. Each trap has a volume of $290cm^3$, holding 110g of charcoal, and samples are collected over 5 minute intervals at a flow rate of approximately 20-30 L min^{-1}. Instrument dimensions are 760 x 300 x 200 mm, weight 20kg and power consumption about 100W.

A small control unit is mounted in the cockpit, and a PC based software package gives the pilot complete control over the operation of the radon sampler. The radon sampler is small and light enough to be mounted in the under-wing pod of a motorised research glider.

Figure 2: Laboratory apparatus for extraction of radon from airborne sampler charcoal traps and delivery to Lucas cells for counting

Back in the laboratory (Fig. 2), exposed charcoal traps are heated to 350°C in a purpose built oven and flushed with nitrogen into a second, smaller charcoal trap kept at low temperature. The second charcoal trap is then heated and flushed with nitrogen to carry the captured radon to one of 20 evacuated Lucas cells for up to 9 hours of α-decay counting. In this way, the charcoal traps can be processed at a sustained rate of one per 90min. Immediately prior to their next exposure, the charcoal traps are "cleaned" of radon by flushing with nitrogen at 350°C for 20 minutes. Flushing continues while the traps are cooled and then capped ready for immediate deployment.

Of the two important errors affecting radon concentration estimates the one caused by the counting system is relatively small. This error, measured as the percentage of net counts, for 10 mBq m^{-3} is less than 30% for 9 hours of counting. The major source of error is caused by small amounts of radium-226 (radon parent element) which are normally present in commercial charcoal. The amount can vary widely from batch to batch. Even if this amount and its contribution to the total count were precisely known, the resulting error after 72 hours of in-growth time at 10 mBq m^{-3} of radon would be 120% and 370% for the two different charcoal materials used in the present study.

2.2 Field measurements

Radon and meteorological profiles were collected in daytime boundary layers during three two-week airborne field campaigns over rural inland Australia. Based out of Goulburn airport (34°48.4′S, 149°44.2′E), we conducted twice-daily flights (weather permitting) over the centre of a broad shallow valley in the Southern Tablelands of New South Wales (elevation 600-700m ASL). The terrain under the flight pattern was relatively flat, dry and homogeneous, being used mainly as grazing pasture in this low-rainfall region.

The sampling pattern consisted of an ascent (to achieve position and altitude, and to identify important layers and levels, including the mixed layer depth, cloud base and cloud top), followed by a simple descending vertical stack of straight-and-level sampling runs each of 5-10min duration, spaced to include: the "clean" air at altitude, the air at mid-cloud level, levels just above and below the mixed layer top (or cloud base), the middle of the mixed layer (sub-cloud layer) and always one run very close to the surface (usually about 30m AGL).

The charcoal traps were prepared (flushed at 350°C for 20 minutes) on-site at Goulburn airport prior to each flight, and then inserted on the aircraft sampler. After landing, they were removed and immediately driven to ANSTO for the laboratory analysis. This resulted in a delay of only about 4 hours between exposure and laboratory processing.

Figure 3: ECO-Dimona instrumented motorized glider operated by *Airborne Research Australia*

Flights were conducted using an instrumented ECO-Dimona motorised glider (Fig. 3) operated by *Airborne Research Australia*, a research group attached to Flinders University of South Australia. The Dimona simultaneously recorded numerous meteorological and navigational quantities to supplement the radon sampling, including high frequency turbulence data suitable for eddy covariance measurements.

A 10m mast was also erected at a nearby farm during each experiment, recording continuous meteorological quantities and surface (2m) radon concentrations using an ANSTO 1500 L dual flow loop, two filter detector (Zahorowski *et al.*, 2005).

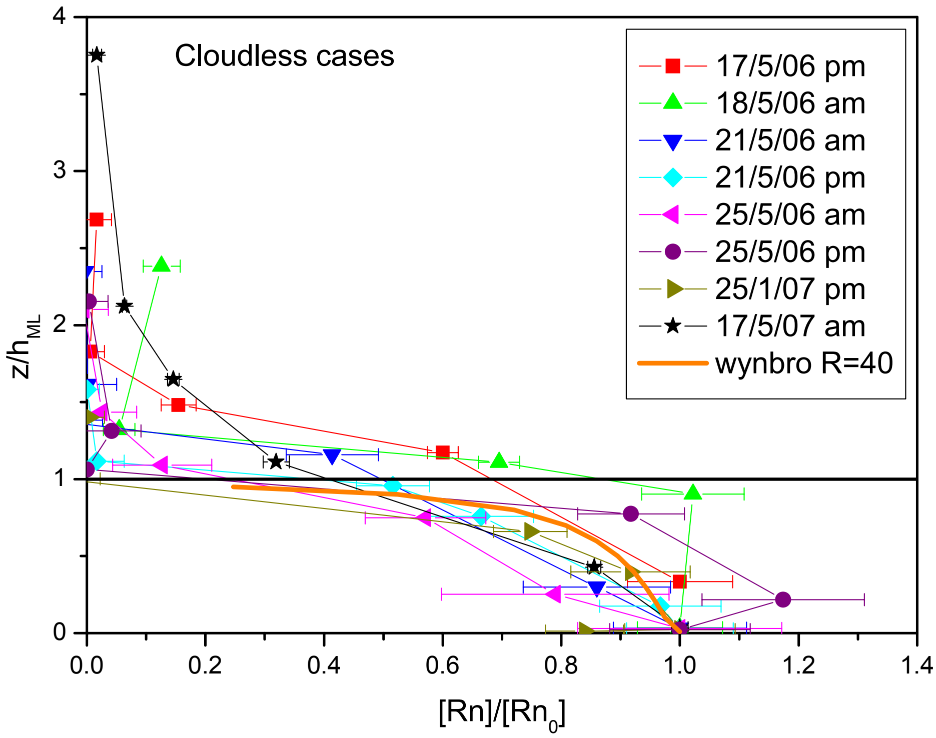

Fig. 4(a)

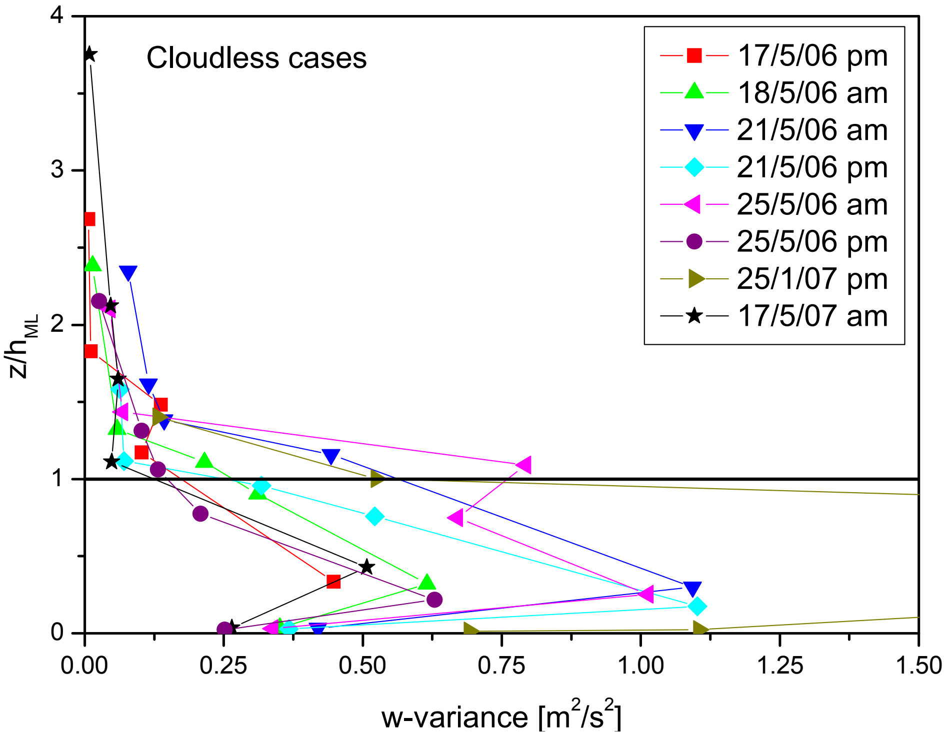

Fig. 4(b)

Figure 4: Profiles of (a) radon normalized with its surface value and (b) vertical velocity variance, for 8 clear-sky convective boundary layers in the Goulburn region of inland rural New South Wales (Australia). Altitude axis is normalized with mixed layer depth (marked with horizontal line). See text for discussion of "wynbro" line

3. RESULTS AND DISCUSSION

We present vertical profiles of radon and meteorological quantities under conditions ranging from clear skies to moderately developed fair-weather cumulus and stratocumulus.

3.1 Clear sky boundary layers

Figure 4 shows profiles of radon concentrations and vertical velocity variances for 8 clear-sky convective boundary layers sampled during the Goulburn campaigns. The radon profiles (Fig. 4a) display a large range of gradients in the upper half of the mixed layer, despite constant potential temperature profiles evident in the soundings in all cases (not shown). Furthermore, the degree of the radon gradient is positively correlated with the strength of the vertical velocity variance in the interior of the mixed layer (Fig. 4b). Indeed, it can be noted that the four cloudless cases with the strongest vertical mixing (light blue, dark blue, brown and magenta) correspond to the four radon profiles with the strongest gradients.

This counter-intuitive behaviour in the radon profiles is a result of "top-down" diffusion in the upper convective boundary layer associated with the entrainment process. Due to its 3.8-day half-life, radon concentrations in the free atmosphere are constrained to be 1-3 orders of magnitude lower than near-surface values. This ensures that a large gradient is maintained between the air in the boundary layer and in the free atmosphere above, leading to significant entrainment of radon across the top of the mixed layer even when the entrainment mass flux is moderate. This underlines the importance of vertical motions in driving entrainment across the boundary layer top, and illuminates the usefulness of radon as an indicator of entrainment in the boundary layer.

The "top-down" diffusion process for scalar mixing in the boundary layer was first investigated in the landmark papers of Wyngaard and Brost (1984) and Moeng and Wyngaard (1984). Theoretical scalar concentration profiles were derived for various values of the ratio R between the entrainment and surface fluxes. R values for temperature and humidity are typically in the range −0.5 to 0.5 (negative for temperature). However, in the case of radon R is expected to be much bigger as the entrainment flux is likely to be large, due to the near-zero radon concentrations above the boundary layer. We demonstrate this in Figure 4(a), where the theoretical radon profile is drawn for R=40 (note that typical surface concentration and flux values from the Goulburn dataset have been used in this calculation). The resultant curve falls approximately in the centre of the range of gradients encountered in the set of cases plotted.

3.2 Cloudy boundary layers

The venting process is further enhanced in the presence of active boundary layer clouds, and radon again proves to be a powerful tracer. In Figure 5, the Goulburn flights are categorized into three degrees of boundary layer cloud conditions:

- *Top graph:* clear skies, or very thin clouds (including thin stratocumulus);
- *Bottom graph:* strongly active non-precipitating cumulus; and
- *Middle graph:* other cloud conditions (mainly developing, dissipating and advected cumulus).

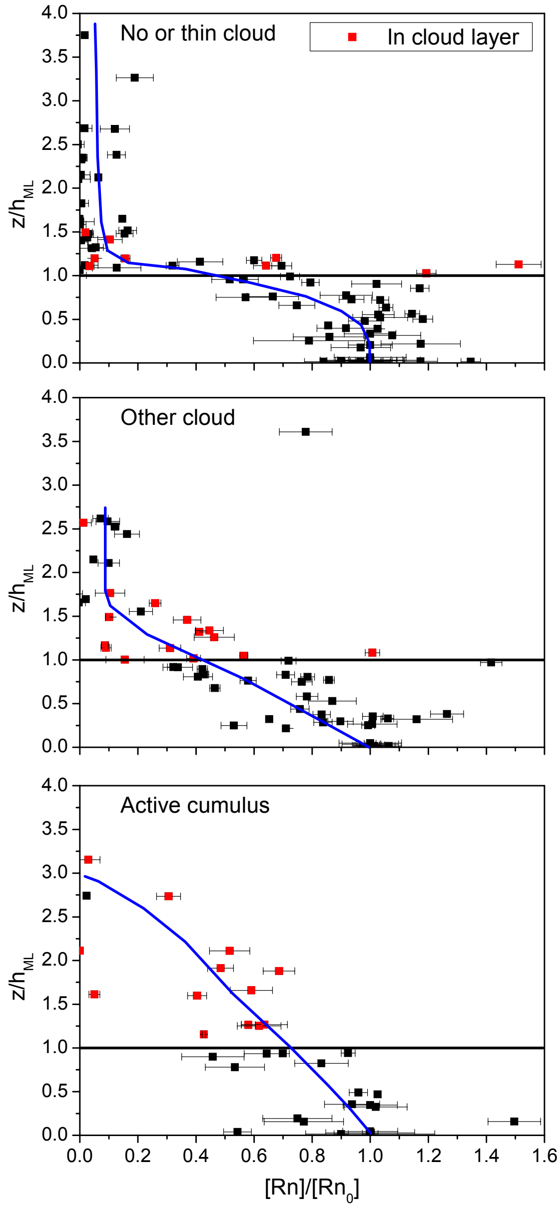

Figure 5: Goulburn radon profiles, arranged into cloud categories (see text). Red points: samples collected in cloud layer. Blue lines drawn by eye

In these graphs, points drawn in red indicate samples collected within the cloud layer. The blue lines, drawn by eye, represent the authors' interpretation of the gross behaviour of the radon profiles.

It can clearly be seen from Figure 5 that when active boundary layer cumulus are present (bottom graph) large radon concentrations are present throughout the cloud layer, and the lack of any discontinuity in the profiles at cloud base indicates that the subcloud and cloud layers are fully coupled. Results in the "other" (middle) category fall neatly between the two extremes, with the "in-cloud-layer" points containing all the high radon values above cloud base (h_{ML}).

Given that the aircraft flew mainly in the spaces between clouds, the measured radon concentrations indicate the extent to which air is being detrained out of the clouds in what is effectively an enhanced boundary layer venting process forced by the action of the clouds.

4. Conclusions

The new ANSTO experimental capability for quantification of atmospheric mixing using airborne radon-222 measurements has been deployed in a series of field campaigns using light aircraft over rural inland Australia. First results from these studies show that radon profile measurements are a quantitative indicator of the degree of vertical mixing and venting in the atmospheric boundary layer under clear and cloudy conditions.

Radon is not well-mixed in the convective boundary layer. Measured profiles under clear-sky convective conditions display significant gradients in the upper mixed layer as a result of "top-down" diffusion processes associated with entrainment.

With active boundary layer cumulus present, large radon concentrations are evident throughout the cloud layer. As the aircraft flew in between the clouds, the measured radon concentrations therefore indicate the extent to which air is being detrained out of the clouds in what is effectively an enhanced boundary layer venting process.

These results demonstrate the usefulness of radon as an unambiguous indicator of mixing and venting in both clear and cloudy boundary layers.

It is hoped that results from these studies will aid in the validation of large eddy simulations and chemical transport models, and ultimately lead to the development of improved parameterisations of vertical transport processes in regional and global climate models.

5. References

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