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7th November 2014

Dr Yves Balkanski
Editor, ACP

Dear Dr Balkanski,

Please find below our response to the third anonymous review of our paper: "On the use of radon for quantifying the effects of atmospheric stability on urban emissions". We would like to thank the reviewer for their constructive feedback and suggestions.

All comments made by the reviewer are addressed individually below.

Kind regards,

Scott Chambers

Responses to specific comments

p25413, line 4 and p25419 line 4: Why is the nocturnal boundary layer shallowest just prior to sunrise? Are there any literatures? Because nocturnal boundary layer may develop as time passes with wind, the depth may not be shallowest near sunrise.

The statement that "the nocturnal boundary layer shallowest just prior to sunrise" is certainly a generalisation, which we will clarify in the revised text. While the characteristics of the nocturnal boundary layer indeed vary considerably from one night to another (through changing wind conditions, intermittent turbulence events, etc., as mentioned in our manuscript and cited literature), on average (over numerous cases), mixing depths reach their minimum value shortly prior to sunrise since the surface has had the longest time (since the previous sunset) to cool, thereby providing the opportunity for the strongest thermal gradient to form in the lowest atmospheric layers – which is responsible for inhibiting near-surface mixing.

p25421, line 28 (red line): There should be some days in which the daily minimum in the daytime is not clear. How did the authors treat such days?

The reference point for the interpolation scheme was always **the minimum** hourly radon concentration recorded between noon (1200h) and 6pm (1800h) each day; there is no ambiguity in this definition. On cases when the afternoon radon concentrations were not very distinct from subsequent nocturnal values (as was sometimes the case for very windy conditions when there was also complete, or almost complete, cloud cover), the interpolation scheme was designed to go from one afternoon (1200-1800h) minimum to the next, by using the minimum number of straight-line segments required to remain less-than-or-equal-to the observed radon concentration.

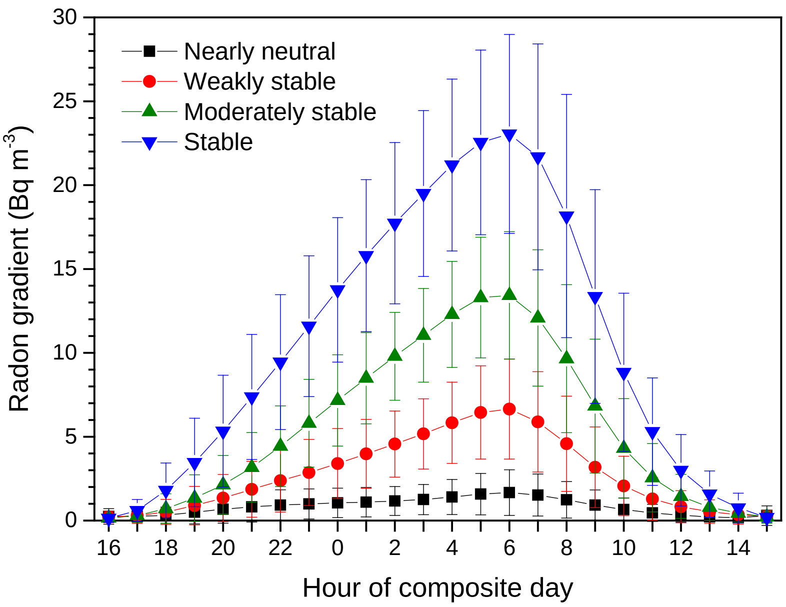
p25423, line 3: The condition for the quartile shown here is for Richmond case. The absolute value of concentration may differ in other places. More generalized criteria should be suggested in addition to the present one.

Indeed, it is correct to say that the absolute radon concentrations of the chosen stability category thresholds will vary from site to site, and that the values pertaining to the stability thresholds at the Richmond site have been shown in the manuscript. However, regardless of the spread of observed absolute radon concentrations at a given site (whether the spread is much greater, or much smaller, than at Richmond), the technique of using quartile boundaries is **completely transferrable** from one site to another. The cumulative frequency diagram (Figure 5) of mean nocturnal radon concentrations within the defined stability window will have a different Y-axis scale for each new site, but this does not prevent new values for the 25th, 50th and 75th percentiles being determined for that site. As such, the quartiles technique is a **completely generalised** approach.

It is also important to appreciate that the choice of quartiles here was **arbitrary**: for a smaller dataset (say, less than a year), we could have defined only **three** stability categories, with boundaries at the 33rd and 66th percentiles; alternatively, for our large (5-year) dataset, it would have been possible to define 10 stability categories (with intervals at each 10th percentile mark), and there would still have been sufficient data for statistically significant results to be generated in each of the 10 categories.

p25423, line 6 Fig.6: Standard deviation of each category may give the information of scattering of the data, which suggest the representativeness of mean value.

We agree that an indication of the scatter within each stability category would be useful – originally we had avoided showing this in case it cluttered the image too much. However, in the revised version of the manuscript we will replace the existing Figure 6 with the version below. Each of the stability categories shown are contributed to by between 410 – 420 whole days of observations.



p25425, line 21: When the authors would like to say about the air pollutants, they should consider the difference of source characteristics from those of radon. For example, spatial distribution and time variation of these sources are much different from radon source. They should add these points to some extent around the measuring point at Richmond.

Thank you for the suggestion. We agree that in the revised manuscript that a more direct comparison between the form of the diurnal curves shown in Figure 6 (for radon) and Figure 8 (for the various pollutants) should be drawn. This would be best described in terms of differences in the spatial and temporal variability in the source/sink terms of the respective quantities. In fact, it is the large difference observed between diurnal characteristics of radon and the other species which has often led to highly variable, or lower-than-expected, correlations between nocturnal radon concentrations, and concentrations of other species in numerous other studies that have attempted to use radon as an indicator of atmospheric stability.

p25425, line 3: I am wondering which types of SO2 sources are dominant near Sydney, high stacks or near surface source. If the dominant source is high stack, the concentration in the daytime is high. On the other hand, if near surface source is dominant, concentration in the daytime becomes low. Fig.8 and Fig.9 suggest the source height is low; however, Fig.11b in winter case suggests the stack height is high (west fetch). Please explain this difference.

It may be a confusing factor that in Figures 8 & 9 the time axis of the figures goes from 1600h – 1500h (to show the whole nocturnal period in one piece), whereas in Figure 11 the time axis of the figures goes from midnight to midnight. When this difference is taken into consideration, all plots indicate that minimum SO_2 concentrations were generally observed at night between 0100 – 0600h.

Regarding Figure 11b, in summer (when flow is mainly from the east) the sources of SO_2 are relatively local (suburban Sydney), and closely track morning / evening peak traffic times (factoring in advection time from Sydney to Richmond). In this respect they represent near-surface sources, but, since they have been advected approximately 30 km prior to reaching Richmond they would be fairly well mixed in the boundary layer, so their characteristics may resemble high stack releases. In winter, however, when regional flow is mainly from the west, SO_2 sources are more distant, and show only a broad increase during the day, decrease at night. Cohen et al., (2012; Atmospheric Environment, 61, 204-211) estimate that 30-50% of sulphate measured in the greater Sydney region at these times can be attributed to releases from distant coal-fired power stations.

The time axis of Figure 11 will be made consistent with that of Figures 8 and 9 in the revised version of the text.

p25426, line 2 Fig.8 and Fig.9: Why different unit is used for pollutants in Fig.8 from Fig.9? The different unit may cause to misleading.

Thank you for the suggestion. The units of Figures 8 & 9 will be changed to match those of Figure 11 in the revised manuscript to avoid confusion.

p25426, line 10 "Comparing ... for the radon scheme. In fact ... days".: The logic of these two sentences are unclear. Do these mean that the P-G method likely classify the case into "D" after sunrise when stable boundary layers still remains under sunshine?

The text in question is referring to the P-G stable (F) and moderately stable (E) categories, not “D”, as mentioned in the comment. The two main points being made are:

1. The average diurnal amplitude of near-surface temperature on “stable” days is less (by 2 degrees) for the PG-classified “stable” days than for the radon-classified “stable” days. This is consistent with (but not irrefutable proof for) the radon technique being more successful at identifying cloud-free, low-wind conditions (usually associated with anti-cyclonic subsidence, and the most stable nocturnal conditions – that give rise to the strongest surface heating on the following mornings); and

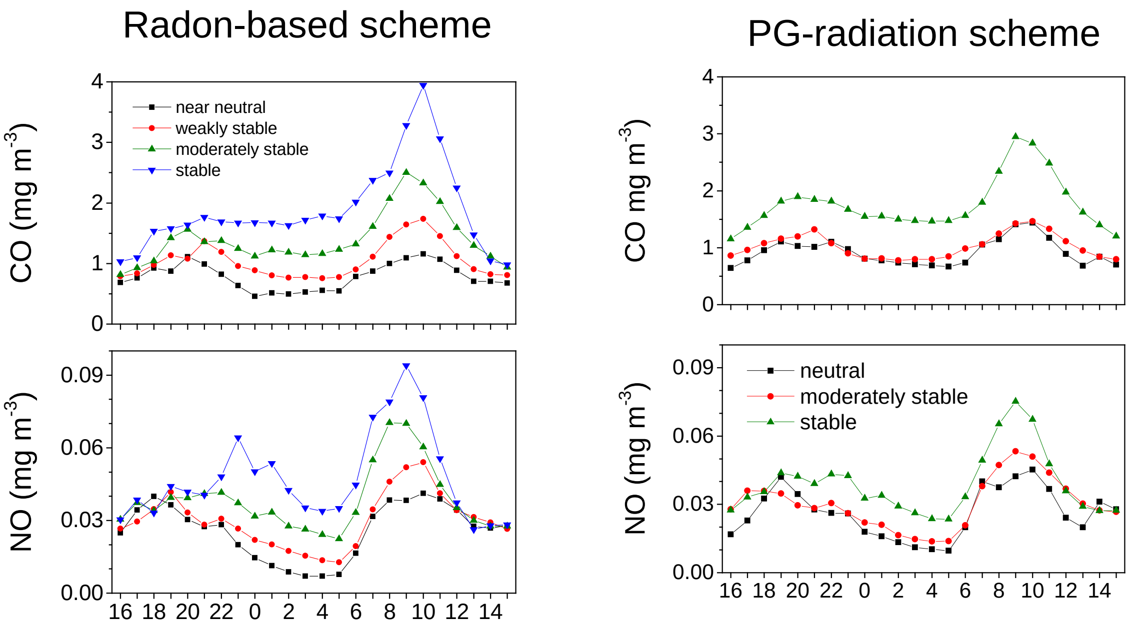
2. The maximum temperatures achieved on the PG-classified “stable” days were, on average, lower than those on the PG-classified “moderately stable” days. Which indicates that the PG-scheme is including some days with hot, clear-sky mornings in the “moderately stable” nocturnal category that might actually have been better suited to the “stable” nocturnal category.

p25428, line 19 "...economical": Is that so? Isn’t the cost of radon monitor expensive?

For nocturnal observations, it can be demonstrated that both the Pasquil-Gifford turbulence stability classification scheme (shown in this manuscript) and PG radiation classification scheme (see example below) are substantially inferior to the radon-based nocturnal stability classification technique. To significantly improve upon the nocturnal stability classification offered by either of the Pasquil-Gifford approaches would require multi-height, research-grade meteorological observations (including high-frequency turbulence observations at each height). A measurement installation of this kind would likely cost of order AUD**$100k**; and require substantial ongoing maintenance to operate effectively. The radon technique, on the other hand, can be employed completely <u>independently of any meteorological observations</u>: i.e. it requires **only a radon detector** in addition to instruments for the pollutant species being observed. Numerous direct and “by progeny” radon detectors (with a wide range of lower-limits-of-detection) are presently available for between: AUD**$15 - $45k** (a substantial upfront saving). Furthermore, at the upper end of this price range (AUD**$45k**), the direct dual-flow-loop, two-filter radon detectors as manufactured at ANSTO, are suited to long-term deployment, and require very little in terms of ongoing maintenance. We believe this justifies the radon-based stability classification technique as comparatively economical.

We provide below a comparison between another application of the radon-based stability classification technique and the Pasquil-Gifford radiation stability classification scheme – an excerpt from a manuscript currently in preparation. Here the radon and PG schemes are used to characterise the influence of stability on CO and NO concentrations in an industrial region of a town in central China. Low wind speeds at this site render the PG-turbulence scheme completely ineffective.

Similarly to the findings of the present paper, here the PG-radiation scheme underestimates peak pollutant concentrations under stable conditions at the industrial site, and poorly distinguishes between “moderately stable” and “neutral” conditions.



p25431 "Summary and conclusions" The method shown here may still be site specific. The remarks for the cases that this method will be applied to other observation site should be added. Because the characteristics of air pollution deeply depend on the distribution of sources of pollutants, I think it is not so easy to apply the radon method developed in this paper to other cities. The authors should show a strategy to implement this method to a variety of cities, if possible.

Section 5.1 of the manuscript addresses some potential limitations and caveats of the method. Most notably, if a site is close to the coast (i.e. within 20km or less of a very strong gradient in the surface radon source function), then considerable care is needed in applying the technique. For example, we have tested the scheme at a site in Wollongong, NSW, that is only 3 km from the coastline. At that site, we had to derive completely separate cumulative frequency diagrams (with different absolute quartile stability thresholds) for onshore vs offshore flow conditions to account for the change in radon source function near the coast. That said, for all other (inland) sites, this method should – in theory – be equally applicable (we have yet to test it in a region where it has not worked).

While the present manuscript applies the technique at an ideal site (inland and flat), the Chinese example mentioned in the previous comment, demonstrates that the technique can be applied equally well to a town in topographically complex valley region that is prone to atmospheric stagnation events. Part of the strength of this technique is that can be derived, and applied, completely independently of the distribution of sources/sinks of the other anthropogenic emissions being investigated: the categorisation scheme is based on the behaviour of radon (a fairly uniformly distributed, unreactive atmospheric tracer), then the days of pollutant observations are subsequently grouped according to these categories. Treated in this way, as evident in Figure 8 of the manuscript, the resultant diurnal cycles of the pollutants clearly show the influences of the spatial/temporal variability in their sources and sinks compared to radon.

An additional caveat that we will mention in the revised manuscript is that at sites where there is either (a) a large seasonal change in the radon source function (due to freezing / snow cover), or (b) a large seasonal change in daylight hours, it may be prudent to define separate cumulative frequency diagrams (Figure 5) for the warmer and cooler halves of the year, since these factors would influence the amount of radon that could accumulate within the defined “stability window” (Figure 4b). Soil freezing and/or snow cover was not a concern at the Richmond site.

The authors would be happy to assist with trialling the implementation of this technique at any site should ambiguities remain in the revised manuscript – please direct any comments/questions to the corresponding author.