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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "The determination of highly time resolved and source separated black carbon emission rates using radon as a tracer of atmospheric dynamics" *by* Asta Gregorič et al.

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Dr. Barbara Ervens, Editor Atmospheric Chemistry and Physics

Dear Dr Ervens,

Thank you for the opportunity to review the discussion paper ACP-2019-911 "The determination of highly time resolved and source separated black carbon emission rates using radon as a tracer of atmospheric dynamics" by Gregorič et al., currently under consideration for publication in Atmospheric Chemistry and Physics. The paper is

dedicated to exploring and explaining differences in seasonal and diurnal source apportioned black carbon (BC) emissions between an urban and a semi-rural setting in Slovenia with strongly contrasting topographic settings. A particular focus of the paper involves the combined use of a box model (e.g. Williams et al. 2016; Salzano et al. 2016) and hourly Radon-222 observations to account for atmospheric dilution influences on BC concentrations, thereby enabling seasonal and diurnal estimates to be made of the separate emission fluxes of traffic-related and biomass burning-related BC. To my knowledge, inverting this kind of box model in order to obtain source apportioned BC emission rates has not previously been published.

Given the significance of BC in atmospheric particulate matter, both in terms of its potential climatic and health impacts, I believe that the study would be of interest to the readership of ACP. However, before I could recommend this manuscript for publication there are some fundamental issues pertaining to the analyses that would need to be addressed, potentially including a revision of the intended study scope. Together these changes would constitute major revision. I have noted my key concerns below.

Specific key concerns

1. Measurement heights and site characteristics: Much well-supported literature (including Karstens et al. 2015; ACP, 15, 12845-12865, 10.5194/acp-15-12845-2015), indicates that radon fluxes near Ajdovščina (AJ) are higher than near Ljubljana (LJ); by at least a factor of two. However, average radon (and its diurnal amplitude) reported in this study are higher at LJ than at AJ (Fig.4). This is likely attributable to: (i) a difference in radon sampling height (1m at LJ and 3m at AJ), (ii) the fact that the AJ radon observations were made on sloping ground, and (iii) the relative proximity of AJ to the coast (∼20 km SW) and significant mountain peaks (∼10 km N – NE). The greater measurement height at AJ would reduce observed radon concentrations cf. LJ (particularly at night), and the sloping terrain would contribute to frequent katabatic flow, which deepens the nocturnal boundary layer (further reducing concentrations), and reduces radon build up within the stable nocturnal boundary layer (SNBL) (since, the ultimate source

air for the katabatic flow is the lower troposphere, where radon concentrations can be very low). In support of this hypothesis, compare the spring diurnal cycles between sites (Fig. 4); AJ observations do not exhibit the distinctive pre-dawn radon peak seen at LJ characteristic of undisturbed accumulation in the SNBL.

Two assumptions of the box model employed in this study are: (i) a well-mixed SNBL, and (ii) a uniform radon source function within the region that could influence the model. In reality, concentration profiles within the SNBL exhibit strong gradients. Consequently, making a direct comparative analysis between sites where concentrations are recorded at different heights from the surface (without correcting for this) could introduce significant biases. Furthermore, while the radon source function near LJ would likely be uniform on spatial scales that influenced the model, this is not the case for AJ. The Adriatic coast lies ∼20km SW of AJ (beyond which the radon flux effectively drops to zero). Mountain peaks of >1000m lie ∼10 km N-NE of the site; at night under low to moderate wind speeds (as selected for this study), air would often be drawn from the lower troposphere, within which radon concentrations can also be very low.

In addition to the differences in radon sampling, BC observations at LJ and AJ were made at 4 and 20m a.g.l., respectively, at the primary sites. As mentioned above, at night under stable conditions, irrespective of potential differences in BC source strengths between the sites, or the flushing effect of katabatic flow at AJ, a significant gradient in BC concentrations would be expected in the SNBL between 4 and 20m agl. Even if both sites were on level ground, it would be necessary to estimate and correct for the separate sampling height differences between the sites before attempting a direct comparative analysis (at least at times when the ABL wasn't well mixed). The advective losses of radon in the SNBL at AJ are a separate complicating factor, and may change with wind direction. While this study excludes the highest 20% of wind speeds, all others are treated equally. In the related study of Williams et al. (2016), the atmospheric class typing approach employed selected several groups of mixing conditions each containing relatively consistent/similar wind speed and direction (which reduced the uncertainty of the spatial decay constant estimates).

2. Unsuitable radon flux estimates: As noted by the authors (e.g. P5 L9; P8 L13-14), successful application of this box-model technique, and subsequent accuracy of the BC emission estimates, is contingent upon reliable knowledge of the radon flux at each site and its seasonal variability. On page 12 the authors outline the approach used to estimate the seasonality of radon fluxes at LJ and AJ. Contrary to existing literature, derived radon fluxes were found to be higher at LJ than at AJ. Furthermore, the reported seasonal variability of radon flux at LJ was from 70 - 150 mBq/m2/s, compared with existing literature estimates of 15 - 25 mBq/m2/s, yet the quoted uncertainty of the adopted flux estimation technique was ±15 mBq/m2/s. Clearly, the derived radon fluxes are not appropriate for use in this study, and I would urge the authors to further investigate the cause of this discrepancy in flux estimates.

Radon fluxes were estimated by regressing mixing depths from the box model (using a range of assumed fluxes), against mixing depths from the NOAA-ARL GDAS database. More information about the data selection criteria for these regressions is warranted here (including an example regression plot). Even if only using fair-weather data it would not be appropriate to make these regressions using values across the whole diurnal cycle since (i) the radon / box-model mixing height estimates are most poorly defined for the 3-5 hours in the mid-afternoon when the GDAS data is most representative of "reality", and (ii) nocturnal mixing depths in the GDAS database are worst at night under stable conditions, when the radon / box-model method works best (in fact, the nocturnal GDAS data has a minimum reported value of 250m a.g.l. for nocturnal mixing under stable conditions; which is around a factor of 2 higher than corresponding nocturnal mixing depths predicted by the radon / box-model method). With this in mind, perhaps the mixing depth transition periods (e.g. between 7am and noon) would be best to use (if the resolution of the GDAS record was adequate)?

3. Afternoon box-model mixing depths: When the authors report whole (24-h) diurnal cycles of effective mixing depths based on the radon / box-model approach, further

discussion regarding the uncertainty of the mid-afternoon values is warranted. In my opinion, neither of the cited papers (Allegrini et al. 1994 or Vecchi et al. 2018) provide robust evidence for the efficacy of this mixing depth calculation approach under convective afternoon conditions. As noted by Williams et al. (2016), several hours after the onset of morning convection a number of the necessary assumptions for the box model approach are no longer valid, until convective mixing begins to decay again in the late afternoon. Typically, for 3-5 hours in the mid-afternoon hourly ΔRn values that form the denominator of equations 9, 11 & 12 approach zero (absolute radon concentrations at this time were also often near the instrument's detection limit). In the mid-afternoon of convective days it is not clear that mixing-related influences on ΔRn dominate over advective influences, and depending on the meteorological conditions of the prior several days, radon concentrations in the lower troposphere (that can be entrained to the ABL once the residual layer has been eroded) can vary by 2 orders of magnitude. Applying a low-pass filter to the radon record (with a 4 – 12 hour cut-off; as done in this study and Vecchi et al. 2018) may improve the stability of the box model, but the actual ABL mixing characteristics at this time on a day to day basis are not correctly represented (since the variability being removed by the filtering process is a mixture of instrumental noise and several competing real physical influences). The largest BC ETR fluxes (with the largest uncertainties), are reported at these times (e.g. 2-4pm) for both workdays and Sundays – despite peak Sunday traffic not occurring at this time. Caution should be used when interpreting values at these times as they could bias daily averages.

The authors have sought to evaluate the fidelity of the radon / box-model's mixing depth estimates in two ways: firstly, using lidar observations (Fig. 9a), where results are very encouraging (for the chosen example) – although the comparison period ends around noon (near the time that the problematic afternoon period referred to above begins); and secondly, with vertical BC profiles recovered by drone. However, the chosen method to retrieve mixing depth estimates from the drone profiles appears to give inconsistent results. A visual inspection of Fig. S3 (a) indicates a well-mixed layer that terminates somewhere between 250 – 300m agl, yet the profile analysis method

returns a value of 412m agl. A visual inspection of Fig. S3b suggests an inversion height roughly 250m agl., whereas the chosen analysis method returns an estimate of 181 m agl. Furthermore, the reported uncertainty for these profile-derived mixing depths is ±1-3m, which is clearly unrealistic. If other parameters were retrieved from the drone (e.g. temperature, humidity or wind speed), these might help to improve the accuracy of the estimates.

4. Scope of investigation: Given the measurement complexities at the AJ site, and frequent failure of measurement conditions to satisfy necessary assumptions for application of the box model, if a more accurate estimate of the local radon flux can be made the authors might consider restricting the scope of their analysis of source apportioned BC emission rates to the Ljubljana region? There would still be sufficient interest and novelty in the results of such a study to warrant publication.

As an example of the influences of spatial heterogeneity of the radon flux near AJ, consider the wind speed threshold of ~2.6 m/s set in this study to retain data for analysis. At 2.6 m/s, air masses arriving at the site in the afternoon from the southwest (Adriatic coast) may have radon concentrations of 0.5 – 1.0 Bq/m3 even for relatively shallow daytime mixing depths (~500m agl). On the other hand, air masses arriving during the afternoon from almost any other direction under comparable atmospheric conditions typically have radon concentrations of 5 – 10 Bq/m3. This change alone (unrelated to the ABL mixing depth) is around half the magnitude of the reported amplitude of the radon diurnal cycle.

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