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Atmospheric Measurement Techniques Discussions

Inter-comparison study of atmospheric ^{222}Rn and ^{222}Rn progeny monitors

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Abstract.

The use of the noble gas radon (^{222}Rn) as tracer for different research studies, for example observation-based estimation of greenhouse gas (GHG) fluxes, has led to the need of high-quality ^{222}Rn activity concentration observations with high spatial and temporal resolution. So far a robust metrology chain for these measurements is not yet available.

A 3-month inter-comparison campaign of atmospheric ^{222}Rn and ^{222}Rn progeny monitors based on different measurement techniques was realized during the fall and winter of 2016-2017 to evaluate: i) calibration and correction factors between monitors necessary to harmonize the atmospheric radon observations; and ii) the dependence of each monitor's response in relation to the sampling height, meteorological and atmospheric aerosol conditions.

Results of this study have shown that: i) all monitors were able to reproduce the atmospheric radon variability on daily basis; ii) linear regression fits between the monitors exhibited slopes between 0.62

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and 1.17 and offsets ranging between -0.85 Bq m^{-3} and -0.23 Bq m^{-3} when sampling 2 m above ground level (a.g.l.). Corresponding results at 100 m a.g.l. exhibited slopes of 0.94 and 1.03 with offsets of -0.13 Bq m^{-3} and 0.01 Bq m^{-3}, respectively; iii) no influence of atmospheric temperature and relative humidity on monitor responses was observed for unsaturated conditions; and iv) changes of the ratio between radon progeny and radon monitor responses were observed under very high atmospheric humidity and under very low atmospheric aerosol concentrations. However, a more statistically robust evaluation of these last influences based on a longer dataset should be conducted to improve the harmonization of the data.

Key words: radon, activity concentration, atmosphere, one-filter, two-filters, electrodeposition

1 Introduction

Over continents, the natural radioactive noble gas radon (^{222}Rn) (half-life $T_{1/2}$ = 3.8 days) is continuously generated within the soil because of the decay of radium (^{226}Ra) (Nazaroff and Nero, 1988; Porstendörfer, 1994) and it can then escape into the atmosphere by diffusion, depending on soil characteristics and meteorological conditions (Grossi et al., 2011, Lopez-Coto et al., 2013; Karstens et al., 2015). The global ^{222}Rn source into the atmosphere is mainly restricted to land surfaces (Szegvary et al., 2009; Karstens et al., 2015), with the ^{222}Rn flux from water surfaces considered negligible for most applications (Schery and Huang, 2004).

In recent decades the atmospheric scientific community has been addressing different research topics using ^{222}Rn as a tracer. Examples of such applications include: the improvement of inverse transport models (Hirao et al., 2010), the improvement of chemical transport models (Jacob and Prather, 1990; Chambers et al. 2019a), the study of atmospheric transport and mixing processes within the planetary boundary layer (Zahorowski et al., 2004; Galmarini, 2006; Baskaran, 2011; Chambers et al., 2011, 2019b; Williams et al., 2011, 2013; Vogel et al. 2013; Vargas et al., 2015; Baskaran, 2016), the experimental estimation of greenhouse gas (GHG) fluxes (Levin et al., 1999; 2011; Vogel et al., 2012; Wada et al., 2013; Grossi et al., 2018), and others listed in Grossi et al. (2016).

In light of this, atmospheric ^{222}Rn measurements have been carried out at numerous monitoring stations of GHG concentrations and air quality using three fundamentally different measurement principles: one filter; two filters; and electrostatic deposition (Stockburger and Sittkus, 1966; Polian, 1986; Hopke, 1989; Whittlestone and Zahorowski, 1998; Paatero et al., 1998; Levin et al., 2002). The two most commonly employed measurement systems at European ^{222}Rn monitoring stations are: the dual-flow-loop two-filter monitor (Whittlestone and Zahorowski, 1998; Zahorowski et al. 2004; Chambers et al., 2011, 2014, 2018; Griffith et al., 2016), which samples and measures radon directly, and the one-filter monitors, of which several kinds are in use (e.g. Stockburger and Sittkus, 1966; Polian, 1986; Paatero et al., 1998; Levin et al., 2002), which sample and measure aerosol-bound radon progeny. Finally, a third method is being used at several Spanish atmospheric stations (Vargas et al., 2015; Hernández-Ceballos et al., 2015; Grossi et al., 2016; Frank et al., 2016; Grossi et al., 2018; Gutiérrez-Álvarez et al., 2019). This type of instrument performs a direct measurement of ^{222}Rn and ^{220}Rn (thoron) activity concentrations using electrostatic deposition of ^{218}Po and ^{216}Po, respectively (Hopke, 1989; Grossi et al., 2012).

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The diversity of the three aforementioned measurement techniques could introduce biases or compatibility issues that would limit the comparability of the results obtained by independent studies and the subsequent application of atmospheric radon data for regional-to-global investigations (e.g. Schmithüsen et al., 2017). Thus, a comparative assessment of all the experimental techniques applied for atmospheric ^{222}Rn activity concentration measurements and a harmonization of their datasets is needed, as suggested by the International Atomic Energy Agency (IAEA, 2012).

Xia et al., 2010 carried out a comparison of the response of a dual-flow-loop two-filter detector from the Australian Nuclear Science and Technology Organisation (ANSTO, Whittlestone and Zahorowski 1998) and a one-filter monitor (α/β Monitor P3) manufactured by the Bundesamt für Strahlenschutz, Germany (BfS) (Stockburger and Sittkus, 1966), for atmospheric ^{222}Rn measurements under various meteorological conditions at 2.5 m above ground level (a.g.l.) over one year. Their results showed that both systems followed the same patterns and produced very similar results most of the time, except under specific meteorological conditions such as when precipitation or the proximity of the forest canopy could remove short-lived progeny from the air mass to be measured by the one-filter monitor. However, Xia et al. (2010) did not find a clear relationship between precipitation intensity and the ratio between progeny-derived ^{222}Rn and ^{222}Rn activity concentration to convert the progeny signal to ^{222}Rn activity concentration.

Grossi et al. (2016) presented results from two short (about 7-9 days) comparisons between a one-filter monitor from Heidelberg University (HRM; Levin et al., 2002), and an Atmospheric Radon MONitor (ARMON, Grossi et al., 2012), an electrostatic deposition monitor from the Universitat Politecnica de Catalunya (UPC). The two comparison campaigns were carried out at a coastal and a mountain site, with sampling in both cases from 10 m a.g.l. These comparisons revealed that the responses of both monitors were in agreement except for water saturated atmospheric conditions or periods of rainfall. Again, the quantity of comparison data was not sufficient to confirm any statistical correlation.

Loss of aerosols in the air intake systems can also complicate the derivation of ^{222}Rn activity concentrations from one-filter systems such as the HRM. Levin et al. (2017) carried out an assessment of ^{222}Rn progeny loss in long tubing by laboratory and field experiments. Results of these experiments, for 8.2 mm inner diameter (ID) Decabon tubing, gave an empirical correction function for ^{222}Rn progeny measurements, which enables the correction of measurements for this specific experimental setup (tubing type and diameter, flow rate, aerosol size distribution).

Finally, Schmithüsen et al. (2017) conducted an extensive European-wide ^{222}Rn/^{222}Rn progeny comparison study in order to evaluate the comparative performance of one-filter and two-filter measurement systems, determining potential systematic biases between them, and estimating correction factors that could be applied to harmonize ^{222}Rn activity concentration estimates for their use as a tracer in various atmospheric applications. In this case, the authors employed a reference HRM monitor, which was taken to nine European measurement stations to run for at least one month at each of them. This reference monitor was run in parallel to other one-filter and the two-filter radon monitors operating at each station of interest.

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Although several intercomparison campaigns have been carried out so far, none of them has included simultaneous observations from one-filter, two-filter and electrostatic deposition methods. Here, we present the results of a three-months intercomparison campaign carried out in the fall and winter of 2016-2017 in Gif Sur Yvette (France) where, for the first time, co-located measurements from monitors based on the three measurement principles were included. Two two-filter ^{222}Rn monitors**,** two single-filter ^{222}Rn progeny monitors and an electrodeposition monitor were run simultaneously under different meteorological and aerosol conditions sampling from heights of 2 and 100 m a.g.l.

The main objectives of the present study were to: i) compare the calibration and correction factors between all monitors required to derive harmonized atmospheric radon activity concentrations; and ii) analyze the influence that meteorological and environmental parameters, as well as sampling height, can have on the finally determined ^{222}Rn activity concentration.

In the present manuscript the applied methodology is reported, including a short presentation of the radon/radon progeny monitors participating in the campaigns, the sampling sites and the statistical analysis carried out. Finally, the results of the study are presented and discussed.

2 Methods

In section 2.1 a short description is given of the monitors compared in the experiment, mainly focusing on measurement techniques, instrument calibration and maintenance. The main characteristics of these monitors are then summarized in Table 1. Section 2.2 presents the French atmospheric stations of Orme de Mérisiers (ODM) and Saclay (SAC) where the two phases of the intercomparison campaign were realized. Section 2.3 shortly described the devices used to measure the environmental parameters and the atmospheric aerosol concentration at this previous sites during the experiment. Finally, the statistical analysis applied is described in section 2.3.

2.1 ^{222}Rn and ^{222}Rn progeny monitors

2.1.1 Dual-flow-loop two-filter detectors

The two 1500 L dual-flow-loop two-filter detectors included in this exercise were designed and built at the Australian Nuclear Science and Technology Organisation (ANSTO). This model of detector, which will henceforth be named ANSTO, is based on a previous design by Thomas and Leclare (1970), with some early iterations of the modified design being described by Whittlestone and Zahorowski (1998) and Brunke et al. (2002). The subsequent evolution of two-filter detectors in recent decades, and the current principle of operation, has been described in detail by Williams and Chambers (2016) and Griffiths et al. (2016).

During the measurement campaign ambient air was sampled continuously at a rate of 83 L min^{-1} through a 50 mm ID HDPE inlet tube and a 400 L delay volume to allow decay of the short-lived ^{220}Rn ($T_{1/2}$= 56 s). The air stream then passes through the first filter, which removes all ambient aerosols as well as ^{222}Rn and ^{220}Rn progeny. The filtered sample, now containing only aerosol-free air and ^{222}Rn gas, enters the main delay volume (1500 L) where ^{222}Rn decay produces new progeny. The newly formed ^{218}Po and ^{214}Po are then collected on a second filter and their subsequent α decays are counted with a ZnS

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144 photomultiplier system. Atmospheric ^{222}Rn activity concentrations are then calculated from the α count
145 rate and the flow rate through the chamber.

146 The detection limit of two-filter detectors is directly related to the volume of the main delay chamber. The
147 lower limit of detection of the 1500 L model used in this study was around 0.03 Bq m^{-3}. Under normal
148 operation ANSTO monitors are automatically calibrated in situ every month by injecting radon into the
149 sampling air stream from a well-characterized Pylon ^{226}Ra source (ca. 41 kBq radium at SAC station) for
150 5 hours at a fixed flow rate of ~100 cc min^{-1}. Automatic instrumental background checks, each lasting 24
151 hours, are also performed every 3 months to keep track of long-lived ^{210}Pb accumulation on the detectors
152 second filter (which should be changed every 5 years). Based on a calibration source uncertainty of 4%,
153 coefficient of variability of valid monthly calibrations of 2-6%, and a counting uncertainty of around 2%
154 for radon concentrations ≥1 Bq m^{-3}, the total measurement of 1500 L ANSTO radon detectors is typically
155 8-12%.

156 Two ANSTO monitors were used during this study. As explained later in the text these monitors are
157 permanently running at SAC and ODM stations. No calibration source was available when the ANSTO
158 monitor was installed at the ODM site, so calibration and background information derived prior to
159 transport have been used. The ANSTO monitors have low-maintenance requirements but, due to their
160 dimensions (2.5 – 3m long) it can be challenging to install them at stations with space restrictions. As an
161 alternative to the 1500 L detectors, a 700 L model is also available, which is more portable and has a
162 detection limit of around 0.04 Bq m^{-3}.

163 **2.1.2 One-filter monitors**

164 One-filter detectors measure the decay rates of aerosol-bound ^{222}Rn progeny directly accumulated by air
165 filtration (Schmithüsen et al., 2017). The ^{222}Rn activity concentration is then calculated assuming a
166 constant disequilibrium factor (F_{eq}) for a given site and sampling height between ^{222}Rn and the measured
167 progeny in the sampled air.

168 In the present study two monitors based on this method were used. One was developed at the Institute of
169 Environmental Physics of Heidelberg University, Germany, and is described in detail by Levin et al.
170 (2002). Rosenfeld (2010) describe the most recent version of this monitor for which the electronics, data
171 acquisition, and evaluation hardware and software have been modernized. The HRM measurement is
172 based on α spectrometry of ^{222}Rn daughters attached to atmospheric aerosols collected on a static quartz
173 fiber filter (QMA Ø 47 mm) using a surface barrier detector (Canberra CAM 900 mm^2 active surface).
174 The detection limit of the HRM is about 0.05 Bq m^{-3} at a flow rate of about 20 L min^{-1} with an
175 uncertainty below ±20% for typical continental atmospheric ^{222}Radon levels above 1 Bq m^{-3}. In the
176 Saclay experiment, where air for the HRM was collected via a 100 m Decabon tubing (see below), the
177 line loss correction of Levin et al. (2017) was applied to all data. No loss of aerosol was assumed in the
178 short tubing used at Orme de Mérisiers station. Since one-filter detectors have no need for any delay
179 chambers but use only a compact filter holder with integrated detector and pre-amplifier, the HRM is a
180 small instrument and therefore easily portable. Regarding maintenance requirements, the quartz fiber
181 filter should be changed monthly.

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The second type of one-filter monitor participating in this study was built at the Laboratoire des Sciences du Climat et de l'Environnement, LSCE, France (Polian, 1986; Biraud, 2000; Schmithüsen et al., 2017). Within this manuscript this monitor will be called the LSCE monitor. This monitor uses a moving filter band system, which allows the determination of atmospheric ^{222}Rn activity concentration based on measurements of its progeny ^{218}Po and ^{214}Po. Attached ^{222}Rn progeny are collected on a cellulose filter (Pöllman–Schneider) over a one-hour period at a flow rate of 160 L min^{-1} and after this aerosol sampling period, the loaded filter is moved to the alpha spectrometry for a one hour measurement period by a scintillator from Harshaw Company and photomultiplier from EMI, Electronics Ltd (Biraud, 2000). The minimum detection activity is about 0.01 Bq m^{-3} with an uncertainty of 20%.

Regarding maintenance on regular basis, the LSCE monitor's filter roll has to be changed every three weeks. Automatic detector background is performed every three weeks and counting efficiency is manually tested with an americium source. The instrument is designed to measure radioactive aerosols a few meters above the ground level. An inlet filter is installed to avoid radon daughters on the main filter roll. In addition, the filter also blocks black carbon or dirt deposition when the instrument is installed in urban areas as the flow rate drops below 9 m^3 h^{-1}. The instrument size is about 25 cm high, 40 cm long and 25 cm deep, and it can be easily deployed at a station.

2.1.3 Electrostatic deposition monitor

The Atmospheric Radon Monitor (ARMON) used in this experiment was designed and built at the Institut de Tècniques Energètiques (INTE) of the UPC. The ARMON is a portable instrument based on method C, consisting of alpha spectrometry of positive ions of ^{218}Po electrostatically collected on a detector (Hopke, 1989). A detailed description of the ARMON is presented by Grossi et al. (2012).

Sampled air with a flow rate between 1-2 L min^{-1}, is first filtered to remove ambient ^{222}Rn and ^{220}Rn progeny and then pumped through a ~20 L spherical detection volume uniformly covered internally with silver. Within this volume the newly formed ^{222}Rn and ^{220}Rn progeny, i.e. positive ^{218}Po and ^{216}Po ions, respectively, are electrostatically collected on a Passivated Implanted Planar Silicon (PIPS) detector surface by an electrostatic field inside the spherical volume. An 8 kV potential is applied between the PIPS detector base and the sphere walls. As for the ANSTO detector, the sensitivity of this instrument type depends on the detector volume. The design of the monitor employed in this study allows a minimum detectable activity concentration of about 0.2 Bq m^{-3} (Grossi et al., 2012). The measurement efficiency of the electrodeposition method is reduced due to neutralization of the positive ^{218}Po in recombination with OH$^-$ ions in the sampled air (Hopke, 1989). Consequently, it is necessary to dry the sampled air as much as possible before it enters the detection volume. To this end, a dew point of <-40°C was maintained at both intercomparison sites using a cryocooler.

Each ARMON is calibrated at the INTE-UPC ^{222}Rn chamber (Vargas et al., 2004) under different ^{222}Rn and relative humidity conditions (Grossi et al., 2012). The radon chamber of the INTE-UPC is a 20 m^3 installation, which allows control of the exhalation rate (0-256 Bq min^{-1}) and the ventilation air flow rate (0-100 L min^{-1}). The ^{222}Rn source is a dry powder material containing 2100 kBq ^{226}Ra activity enclosed in the source container (RN-1025 model manufactured by Pylon Electronics). The calibration factor F_{cal} of

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the ARMON used in this study was of 0.39 counts per minute (cpm) per Bq m^{-3} with an uncertainty of 10%. The correction factor for the humidity influence inside the sphere was of $6.5 \cdot 10^{-5}$ per part per million H_2O (ppm) with an maximum uncertainty of 10%. The total uncertainty of the atmospheric radon activity concentration measured by the ARMON is of 20%. Every 1-2 years the progeny filter at the ARMON inlet should be changed.

Monitor	Method	α Spectrum	Flow Rate (L min^{-1})	Detection Limit (Bq m^{-3})	Typical uncertanty	Remote Control	Need of dry air sample	Need of corrections depending on the height of the inlet	Portability Level	References
ANSTO	Dual-flow-loop two-filter	No	~83	0.03	8-12%	Yes	No	No	Low	Whittlestone and Zahorowski (1998); Brunke et al. (2002)
ARMON	Electrostatic deposition	Yes	1-2	~0.2	20%	Yes	Yes	No	Medium	Grossi et al. (2012)
HRM	One-filter	Yes	20	~0.05	15-20%	Yes	No	Yes	High	Levin et al. (2002)
LSCE	One-filter	Yes	160	~0.01	20%	Yes	No	Yes	High	Polian, 1986; Biraud, 2000

Table 1. Summary of principal characteristics of the ^{222}Rn and ^{222}Rn progeny monitors compared in the present study.

2.2 Sites

The intercomparison study was carried out at two stations located 30 km southwest of Paris in the fall and winter of 2016-2017 (Figure 1). Both stations, 3.5 km apart, belong to the LSCE and are located in a region with a radon flux of ca. 5-10 mBq m^{-2} s^{-1} in winter, according to output of the Karsten et al. (2015) model.

Phase 1 of the measurements started at Orme des Mérisiers (ODM, latitude 48.698, longitude 2.146, 167 m above sea level) and ran between 25 November 2016 and 23 January 2017. Here, LSCE and ANSTO (for convenience named here as ANSTO_ODM) monitors are routinely running. During Phase I of the intercomparison exercise these two monitors were operated in parallel with a HRM and an ARMON. The sampling height for all radon detectors at ODM was 2 m a.g.l.

Phase II of the exercise was realized at Saclay (SAC, latitude 48.730, longitude 2.180, Figure 1) between 25 January 2017 and 13 February 2017. At this location the sampling inlet height was at 100 m a.g.l. At SAC station an ANSTO monitor (from now on labelled as ANSTO_SAC) was already running. In addition, during Phase II this detector was running in parallel with the portable ARMON and HRM detectors. The LSCE monitor did not participate in Phase II of the experiment.

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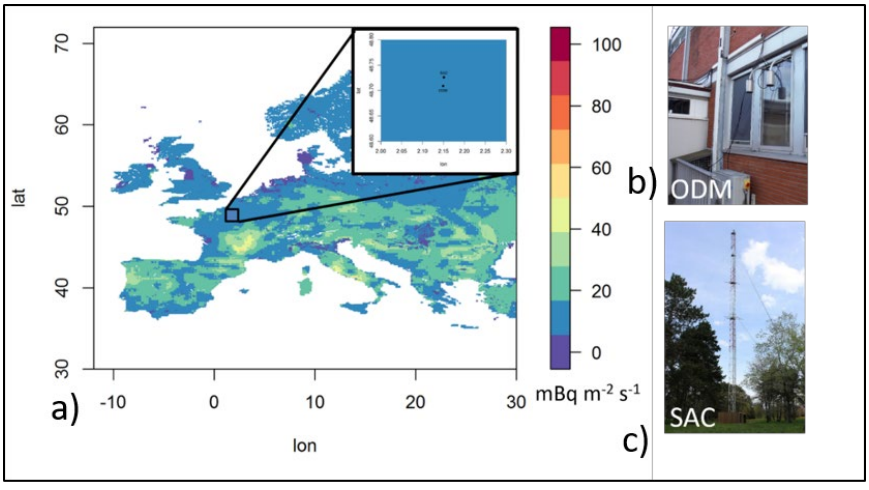
243 Meteorological parameters were also available at both stations during the intercomparison periods at
244 heights corresponding to the radon measurements (2 m and 100 m a.g.l.). In the case of the ODM site,
245 atmospheric aerosol concentrations were also measured for this period.



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247 Figure 1. The INGOSv2.0 ^{222}Rn flux map (Karstens et al., 2015) is shown for a typical winter month
248 (December), with locations of the ODM and SAC sites shown in the inset (a). The radon sampling inlets
249 are shown both for ODM (b) and SAC (c).

250 **2.3 Environmental parameters and atmospheric aerosol concentration**

251 Meteorological data used within this study were variables because continuously measured at the SAC and
252 ODM stations at different heights. The measurements are carried out with a Vaisala Weather Transmitter
253 WXT520 (Campbell Scientific) for: (1) wind speed and direction (accuracies of ± 3 % and ± 3 ºC,
254 respectively); (2) Humidity and temperature (accuracies of ± 3 % and ± 0.3 ºC, respectively). In addition,
255 the atmospheric aerosol concentration is measured at ODM site using a Fine dust measurement device
256 Fidas® 200 S (Palas). The measurement range is between 0 and 20.000 particles cm^{-3}. All the accuracies
257 refer to the manufacturer's specifications.
258

259 **2.4 Data Analysis**

260 **2.4.1 Correlation factors between monitors**

261 In order to study the correlation between responses of the different detectors, linear regression models
262 were calculated using hourly atmospheric radon activity concentrations from each monitor. The linear
263 regression fits were calculated following Krystek and Anton (2007), relative to the two portable detectors,
264 ARMON and HRM, because they both were measuring at SAC and at ODM.
265

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2.4.2 Analysis of the influence of the environmental and meteorological parameters on detector response

The present study intended to build upon the findings of Xia et al. (2010) and Schmithüsen et al., (2017) regarding the possible influence of meteorological conditions on the response of radon and radon progeny monitors.

With this in mind, the ratio between hourly atmospheric ^{222}Rn activity concentrations measured and/or obtained by the HRM, LSCE and ANSTO monitors, and that measured by the ARMON were calculated, and their variability analyzed in relation to hourly atmospheric temperature, relative humidity and atmospheric aerosol concentration measured at ODM and at SAC, respectively. For this part of the study, the ARMON was used as reference being the only portable direct radon monitor running at both sites.

3 Results

Hourly time series of atmospheric ^{222}Rn, in the case of ARMON and ANSTO monitors, and ^{222}Rn progeny (^{214}Po activity concentration) for the HRM and LSCE monitors, measured at ODM and SAC during Phase I and Phase II of the intercomparison experiment are presented in Figures 2 and 3, respectively. In each of the previous Figures, a zoom plot has been also reported as example to look at the response of each monitor to the sub-diurnal atmospheric radon variability. As shown, all monitors running at both sites follow this variability, with ^{222}Rn and ^{222}Rn progeny data measured or estimated by the three different measurement techniques showing the same general patterns. Table 2 summaries the means, minima and maxima hourly atmospheric radon or radon progeny activity concentrations measured by each monitor for both campaigns.

3.1 Phase I: ODM site

During Phase I the LSCE, HRM, ARMON and ANSTO_ODM monitors were operating in parallel, sampling air from the same height (2 m a.g.l.). The mean temperature over Phase I of the campaign was 2.9 ºC with an interquartile range of 0.10 ºC to 5.8 ºC. The mean relative humidity was 80% with an interquartile range of 73% to 89%. An average accumulated rain per day of 13 mm was recorded. The main wind patterns during Phase I were from northeast and southwest, with speeds typically between 1 and 7 m s^{-1}. The mean atmospheric aerosol concentration observed at ODM during Phase I was 505 particles cm^{-3} with an interquartile range of 233 cm^{-3} to 660 cm^{-3}.

The means of the atmospheric ^{222}Rn activity concentration measured by the ARMON and the ANSTO_ODM are in the same order (Table 2). The means of the atmospheric radon daughter activity concentrations measured by LSCE monitor is ca. 50% lower and by the HRM is ca. 30% lower than the atmospheric ^{222}Rn activity concentration.

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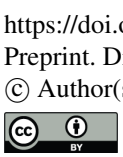

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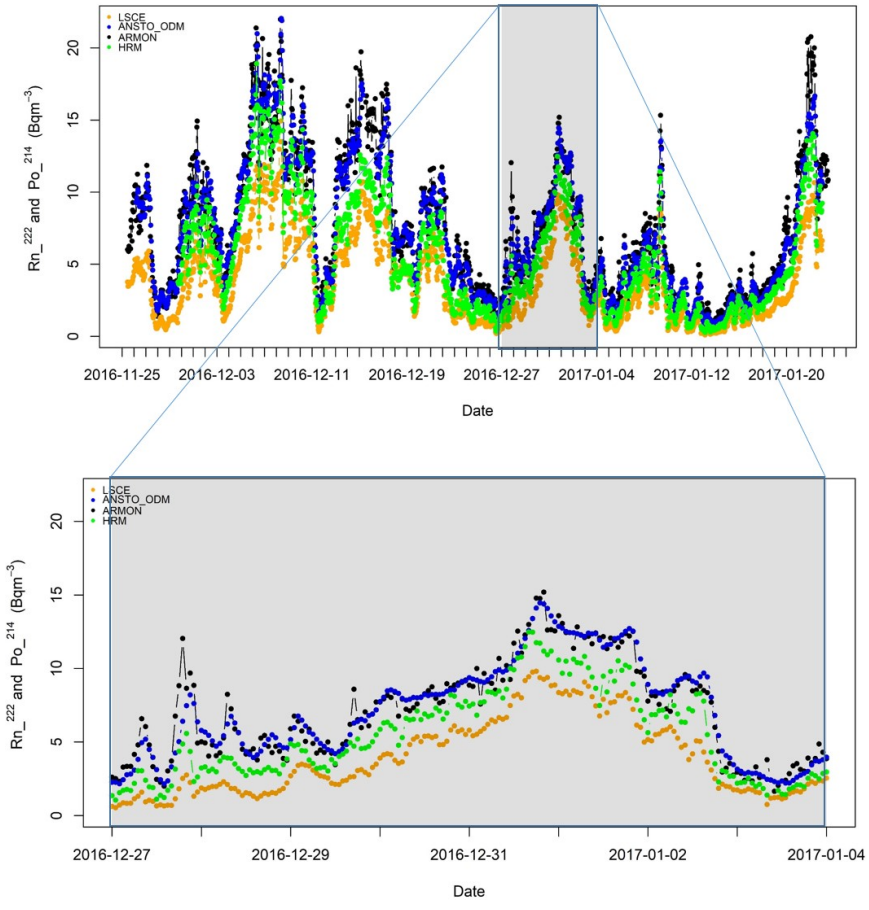

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299

300 Figure 2. Main panel: Hourly time series of the atmospheric ^{222}Rn and, in the case of LSCE and HRM
301 data ^{214}Po activity concentration measured at Orme de Merisiers (ODM) station during Phase I (between
302 25 November 2016 and 23 January 2017) by: ARMON (black circles), ANSTO_ODM (blue circles),
303 HRM (green circles) and LSCE (orange circles) monitors. Zoomed panel: Hourly time series of the
304 atmospheric ^{222}Rn and ^{214}Po measured between 27th December 2016 and 04th January 2017.

305 Table 2 shows the slopes (*b*) and intercepts (*a*) of the linear regression fits calculated between the hourly
306 atmospheric ^{222}Rn and ^{214}Po activity concentrations measured by the ARMON and/or the HRM and the
307 other ^{222}Rn and ^{222}Rn progeny monitors deployed in Phase I. The calculated slopes were in the range of
308 0.62 to 1.17 and the R^2 values varied between 0.90 and 0.96. The slope closest to unity was calculated
309 between the ARMON and ANSTO_ODM monitors, and was 0.96±0.01, while the lowest slope was
310 observed between the ARMON and LSCE monitors, and was 0.62±0.01. The highest correlation
311 (R^2=0.96) was found between the HRM and LSCE monitors. The plots of the linear regression fits of the
312 Phase I are shown in the Figures S1, S2 and S3 left panel of the supporting material. Notably, the offset (*a*

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313 value) of the regression between the ANSTO and ARMON detectors at ODM is considerably greater than
314 that at SAC. The regression slopes are also slightly different, though not significantly so. These
315 differences are likely related to the limited calibration and background information available for the
316 ANSTO_ODM detector for this intercomparison project. In particular, a substantial component of the
317 instrumental background signal is site specific. This is likely responsible for much of the change in offset
318 value.

				x					
y	**Monitors Phase I**	**Mean (Bq m^{-3})**	**Min/Max (Bq m^{-3})**	***b* (ARMON)**	***a* (ARMON)**	**R^2 (ARMON)**	***b* (HRM)**	***a* (HRM)**	**R^2 (HRM)**
	ANSTO_ODM	7.02	0.73/22.04	0.96±0.01	-0.23±0.03	0.94	1.17±0.01	0.63±0.03	0.93
	HRM	5.45	0.26/18.91	0.82±0.01	-0.71±0.03	0.93	-	-	-
	ARMON	7.55	0.50/21.98	-	-	-	-	-	-
	LSCE	3.84	0.10/14.93	0.62±0.01	-0.85±0.03	0.90	0.76±0.004	-0.29±0.03	0.96
	Monitors Phase II	**Mean (Bq m^{-3})**	**Min/Max (Bq m^{-3})**	**Slope (ARMON)**	**Intercept (ARMON)**	**R^2 (ARMON)**	**Slope (HRM)**	**Intercept (HRM)**	**R^2 (HRM)**
	ANSTO_SAC	3.50	0.43/10.71	0.97±0.01	0.01±0.06	0.95	1.03±0.01	0.15±0.06	0.90
	HRM	3.26	0.26/11.15	0.94±0.01	-0.13±0.06	0.91	-	-	-
	ARMON	3.60	0.17/11.51	-	-	-	-	-	-

319

320 Table 2. The means, maxima, and minima of the atmospheric ^{222}Rn and ^{214}Po activity concentration
321 observed by each monitor participating in the Phase I and II of the intercomparison campaigns. In
322 addition are here reported the slopes (*b*) and intercepts (*a*) of the linear regression fits calculated between
323 the hourly atmospheric ^{222}Rn and ^{214}Po activity concentrations measured by the ARMON and/or the HRM
324 and the other ^{222}Rn and ^{222}Rn progeny monitors deployed in both phases.

325 **3.2 Phase II: SAC station**

326 Phase II lasted 18 days. The mean temperature during this period was 5 ºC with an interquartile range of 2
327 ºC to 8 ºC. The mean relative humidity was 86% with an interquartile range of 80% to 94%. An average
328 accumulated rain per day of 3 mm was recorded. The main wind patterns during this phase at 100 m a.g.l.
329 were from the south and southwest with speeds typically between 3 and 10 m s^{-1}.

330 Figure 3 shows the hourly atmospheric ^{222}Rn and ^{214}Po activity concentrations observed at SAC during
331 Phase II by the ARMON, HRM and ANSTO_SAC instruments. Table 2 reports the means, minima, and
332 maxima of the atmospheric data measured during Phase II by all participating monitors. In this case, the
333 mean atmospheric ^{222}Rn and ^{214}Po activity concentrations measured by all monitors agreed within the
334 instruments errors. At 100 m a.g.l. the slopes of the hourly fits of the monitor's response in this case were
335 all close to unity. The calculated offsets also decreased at 100 m a.g.l. relative to 2 m a.g.l. The plots of
336 the linear regression fits of Phase II are shown in the Figures S3 right panel and S4 of the supporting
337 material.

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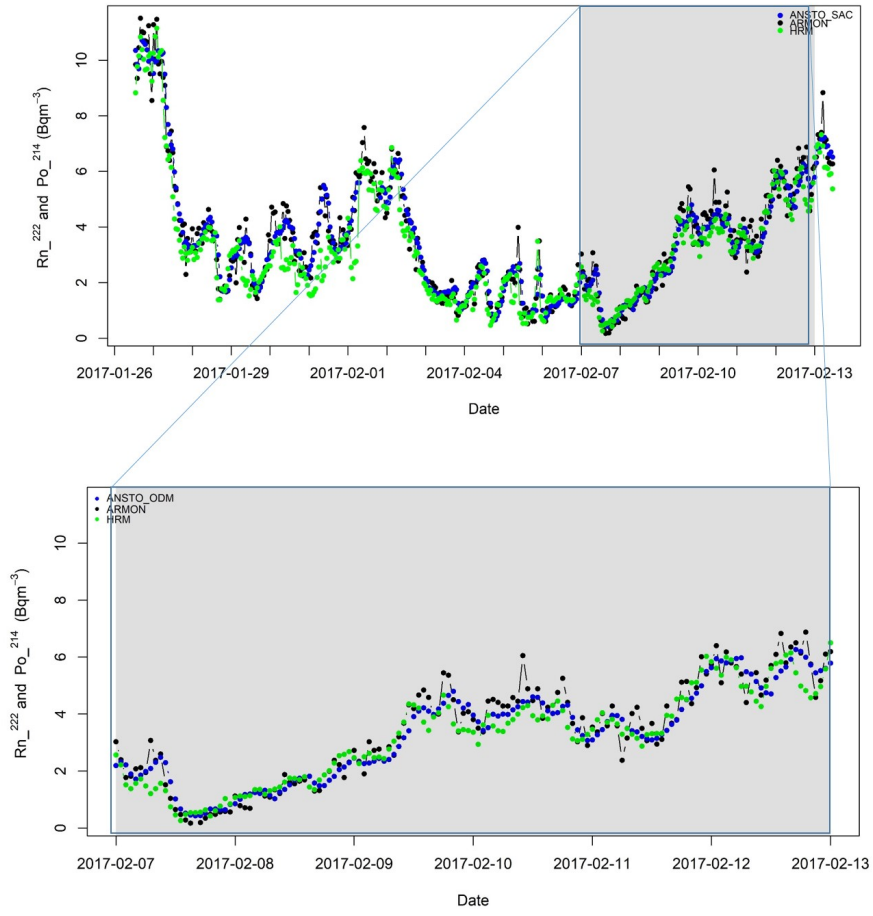

Figure 3. Main panel: Hourly time series of the atmospheric ^{222}Rn and ^{214}Po (HRM) activity concentration measured at Saclay (SAC) station between 25 January 2017 and 13 February 2017 by: ARMON (black circles), ANSTO_SAC (blue circles) and HRM (green circles) monitors. Zoomed panel: Hourly time series of the atmospheric ^{222}Rn and ^{214}Po measured between 7 February 2017 and 13 February 2017.

Figure 2 and 3 show a larger hourly variability of the HRM and ARMON signals compared with the ANSTO ones. This difference in variability is attributable to the combination of a larger counting uncertainty of the HRM and ARMON detectors, and that only an approximated response time correction could be applied to the output of the ANSTO detectors (Griffiths et al. 2016). Further investigations should be carried out to clarify these differences and to exactly quantify the detectors uncertainties for the low ^{222}Rn concentrations typical for outdoor environmental monitoring at or above 100 m a.g.l. During the period of Jan 30 – February 1, 2019, the HRM shows significantly lower values than the ANSTO and ARMON. This period coincides with saturated air humidity conditions.

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3.3 Influence of the weather conditions on the ratio between direct ^{222}Rn and ^{214}Po measurements

Figure 4 and 5 show the variability of the ratio between hourly atmospheric ^{222}Rn and/or ^{214}Po activity concentration measured by each monitor relative to those measured by the ARMON at ODM (Figure 4, upper panels) and at SAC (Figure 4, bottom panels) and by the ANSTO_ODM at ODM (Figure 5, upper panels) and by the ANSTO_SAC at SAC (Figure 5, bottom panels) versus the hourly means of ambient temperature (Figures 4 and 5, left panels) and relative humidity (Figures 4 and 5, right panels) measured at the corresponding stations. Data does not show any evident patterns, which could indicate that there is any impact on ^{222}Rn or ^{222}Rn progeny measurements due to change of ambient temperature and relative humidity, at least not until saturated conditions are achieved. Looking at Figure 5, there appears to be less scatter in the point clouds (particularly at SAC) when the ANSTO_SAC monitor is used as the reference, likely attributable to the lower measurement uncertainty of the ANSTO monitors.

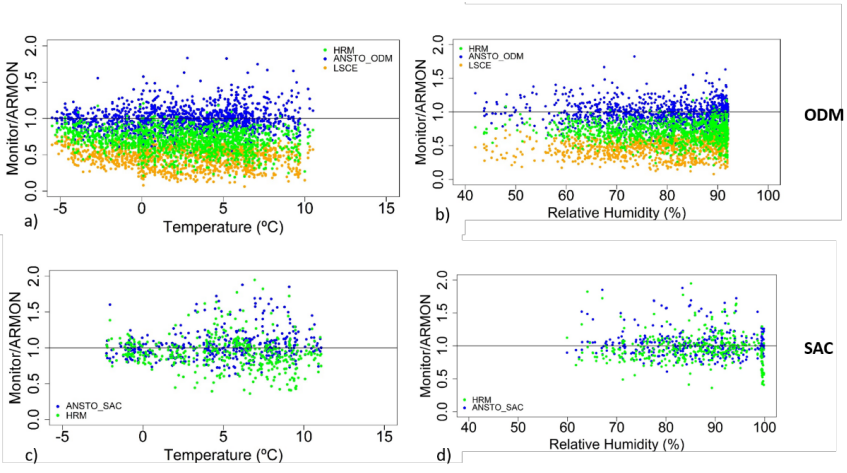

Figure 4. Hourly atmospheric ^{222}Rn or ^{214}Po activity concentration obtained by HRM, LSCE and ANSTO monitors divided by the ^{222}Rn activity concentration measured by the ARMON detector as function of the hourly measured atmospheric temperature and relative humidity at ODM (a and b) and at SAC (c and d), respectively.

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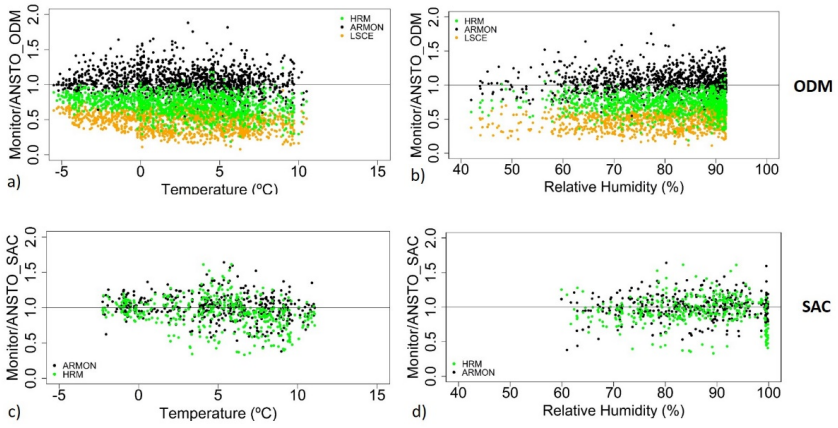

Figure 5. Hourly atmospheric ^{222}Rn or ^{214}Po activity concentration obtained by ARMON, HRM and LSCE monitors divided by the ^{222}Rn activity concentration measured by the ANSTO detectors as function of the hourly measured atmospheric temperature and relative humidity at ODM (a and b) and at SAC (c and d), respectively.

In Figure 6 the ratio of the hourly atmospheric ^{222}Rn or ^{222}Rn progeny activity concentration measured by the HRM (^{214}Po in Figure 6a), the LSCE (^{214}Po in Figure 6b) and the ANSTO_ODM (^{222}Rn in Figure 6c) monitor and the ^{222}Rn activity concentration measured with ARMON (^{222}Rn) are plotted against the logarithm of the hourly aerosol concentration data. Data indicate the existence of a linear relationship between these variables, i.e. of the form:

$$\frac{^{222}Rn\,(Monitor\_i)}{^{222}Rn\,(ARMON)} = a + b \cdot Log_{10}(Aerosol\ Conc.). \quad (1)$$

Here $^{222}Rn\ (Monitor\_i)$ is the hourly atmospheric ^{222}Rn or ^{214}Po activity concentration measured by individual monitors HRM (^{214}Po), LSCE (^{214}Po) and ANSTO_ODM (^{222}Rn), $^{222}Rn\ (ARMON)$ is the one measured by the ARMON monitor and *Aerosol Conc*. is the hourly atmospheric aerosol concentration measured at ODM during Phase I. The results of the linear regression fits for each compared monitor are reported in Table 3. The slope of the ratio between the ANSTO_ODM and ARMON monitors in relation to the variability of the logarithm of the hourly atmospheric aerosol concentration is close to zero and the intercept is close to one. The ratio between the hourly atmospheric aerosol-bound radon progeny data measured by the two one-filter radon progeny monitors and the one measured by the ARMON seems to decrease with decreasing aerosol concentration (Figures 6a and 6b). However, this effect becomes only evident when atmospheric aerosol concentration is lower than 300 particles cm^3.

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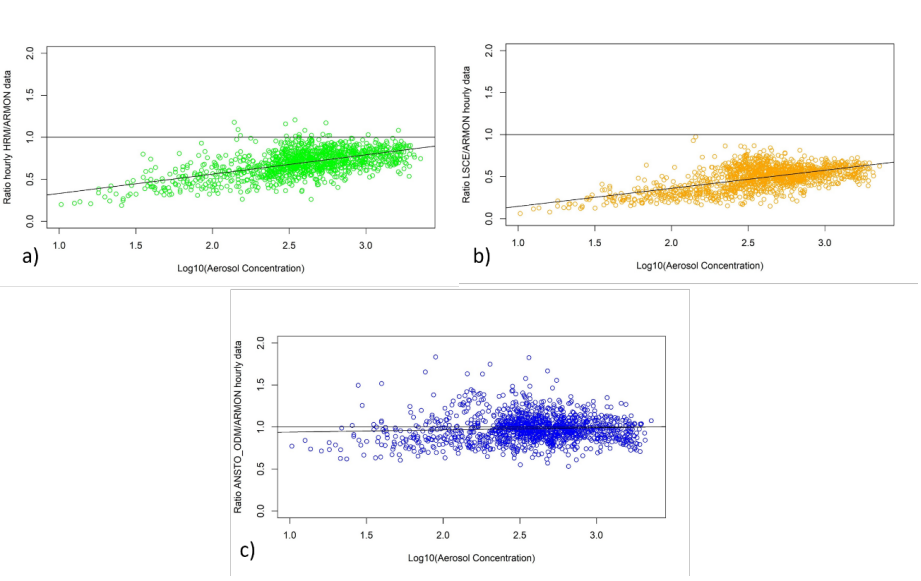

Figure 6. Ratio of the atmospheric ^{222}Rn or ^{214}Po activity concentration measured by the HRM (green dots), LSCE (orange dots) and ANSTO_ODM (blue dots) monitors and those measured by the reference ARMON monitor against the logarithm of the atmospheric aerosol concentration measured at ODM station.

Monitor	*a*	*b*	R^2
HRM	0.10±0.02	0.23±0.01	0.34
LSCE	-0.07±0.02	0.21±0.01	0.34
ANSTO_ODM	0.91±0.03	0.03±0.01	$0.04 \cdot 10^{-1}$

Table 3. Intercepts and slopes of the linear regression fits of the Equation 1

Conclusions

In order to confirm and build upon the results obtained by Xia et al. (2010), Grossi et al. (2016) and Schmithüsen et al. (2017) a three months intercomparison campaign was carried out in the south of Paris, France, in the fall-winter period of 2016-2017. For the first time, three fundamentally distinct radon and radon progeny measurement approaches deployed at GHG observation sites across Europe were compared side-by-side at two measurement heights: 2 and 100 m a.g.l., under a range of environmental conditions with the aim to compare their responses under various atmospheric and/or meteorological conditions.

The results of this study show that ^{222}Rn and ^{222}Rn progeny measurements follow the same general patterns of diurnal variability, both close to and further up from the surface. The slopes and intercepts of the linear regression fits between the direct radon and the radon-progeny measurements are not significantly different from one at 100m height above ground (SAC), but they differ at the 2m level (ODM). This behavior is attributable to the disequilibrium known to exist between ^{222}Rn freshly emitted from the ground and its short-lived progeny in the lowest 10s of meters of the atmosphere, the magnitude

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of which is known to decrease with distance from the surface, as shown in earlier work, and to be close to one at a height of 100m and above (e.g. Jacobi and André, 1963; Schmithüsen et al., 2017).

For the 2 m level, we found a very good correlation of radon progeny activity concentrations between LSCE and HRM measurements (see Figure S1 in the Supplement). The slope, however, is only equal to 0.76±0.04. This number is slightly larger but within uncertainties well comparable to the number reported by Schmithüsen et al. (2017) of 0.68±0.03 (see their Table 2) based on a comparison campaign conducted at ODM in March and April 2014.

Observations of the total atmospheric aerosol concentration available at ODM station during the first two months of the experiment were used to investigate the influence of changing atmospheric aerosol concentrations on the response of the radon/radon progeny measurements. Under very low atmospheric aerosol burden (< 300 particles cm^{-3}), the ^{222}Rn progeny monitors seem to underestimate the atmospheric ^{214}Po activity concentrations by up to 50%. This effect may be attributable to loss of un-attached ^{218}Po and ^{214}Po. Particle number concentrations below 300 particles cm^{-3} at continental stations are, however, very rare and even during winter at Alpine stations like Schneefernerhaus such low particle concentrations are only occasionally observed (Birmili et al., 2009).

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