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Characterising fifteen years of continuous atmospheric radon activity observations at Cape Point (South Africa)

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ABSTRACT

This paper describes and discusses fifteen years (1999–2013) of continuous hourly atmospheric radon (^{222}Rn) monitoring at the coastal low-altitude Southern Hemisphere Cape Point Station in South Africa. A strong seasonal cycle is evident in the observed radon concentrations, with maxima during the winter months, when air masses arriving at the Cape Point station from over the African continental surface are more frequently observed, and minima during the summer months, when an oceanic fetch is predominant. An atmospheric mean radon activity concentration of 676 ± 2 mBq/m^3 is found over the 15-year record, having a strongly skewed distribution that exhibits a large number of events falling into a compact range of low values (corresponding to oceanic air masses), and a smaller number of events with high radon values spread over a wide range (corresponding to continental air masses). The mean radon concentration from continental air masses (1 004 ± 6 mBq/m^3) is about two times higher compared to oceanic air masses (479 ± 3 mBq/m^3). The number of atmospheric radon events observed is strongly dependent on the wind direction. A power spectral Fast Fourier Transform analysis of the 15-year radon time series reveals prominent peaks at semi-diurnal, diurnal and annual timescales. Two inter-annual radon periodicities have been established, the diurnal 0.98 ± 0.04 day^{-1} and half-diurnal 2.07 ± 0.15 day^{-1}. The annual peak reflects major seasonal changes in the patterns of offshore versus onshore flow associated with regional/hemispheric circulation patterns, whereas the diurnal and semi-diurnal peaks together reflect the influence of local nocturnal radon build-up over land, and the interplay between mesoscale sea/land breezes. The winter-time diurnal radon concentration had a significant decrease of about 200 mBq/m^3 (17%) while the summer-time diurnal radon concentration revealed nearly no changes. A slow decline in the higher radon percentiles (75th and 95th) for the winter and spring seasons is found over the 15-year data set, with most of the change occurring in the first 9 years (1999–2007). This observed inter-annual decline appears to be associated with changes in the frequency of air masses having originated from over the African continental surfaces, and no significant trend is found in the lower radon percentiles associated with oceanic air masses. The general decrease of atmospheric radon-associated with continental air-masses at Cape Point could be attributed to changing meteorological conditions, possibly driven by climate change.

1. Introduction

^{222}Rn (hereafter referred to as radon) is a naturally occurring noble gas, mostly of continental origin, frequently used for atmospheric tracer studies (Zahorowski et al., 2004; Crawford et al., 2009; Chambers et al., 2011; Williams et al.,2011). Radon is a radioactive α-decay product of long-lived ^{226}Ra ($t_{1/2}$ = 1 600 y), ubiquitous in most rock and soil types (Botha et al., 2017). Radon has a relatively consistent flux from ice-free terrestrial surfaces of 15–25 mBq m^{-2} s^{-1} varying mainly with soil characteristics such as composition, porosity, moisture and permeability (Ball et al., 1991; Griffiths et al., 2010; Karstens et al., 2015). The radon flux density from ice-free marine surfaces is two to three orders of magnitude smaller than that of continental regions (Schery and Huang, 2004; Zahorowski et al., 2013). As a result of these source

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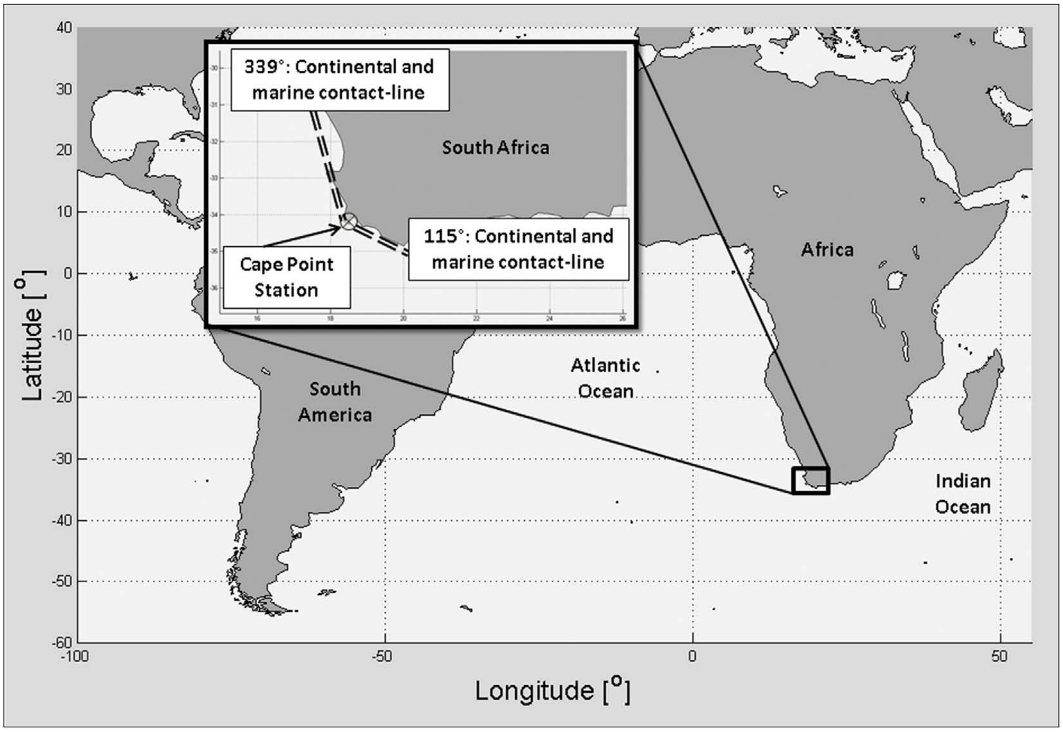

Fig. 1. Map indicates the location of the Cape Point Station. Two directional lines were inserted to indicate contact zones between continental and marine air masses regions.

characteristics, together with its unreactive nature and a radioactive half-life of 3.82 days, radon is an effective natural tracer by which continental air can be distinguished from marine air due to large activity concentration differences existing between the two air mass regimes (Brunke et al., 2004; Chambers et al., 2016).

At coastal baseline stations such as the Cape Point Global Atmospheric Watch (GAW) Station in South Africa, both continental and marine air masses are experienced (Fig. 1). Atmospheric radon measurements at Cape Point (CPT) have been utilised in various studies, including air-mass origin characterisation (Brunke et al., 2004); CO_2 source region identification for assessment via modelling studies; ^{222}Rn and meteorological data correlation studies (Brunke et al., 2004); and the characterisation of trace-gas source regions (Whittlestone et al., 2009). In this paper, we summarize the characteristics of, and trends in, a newly compiled fifteen-year data set (1999–2013) of continuous hourly atmospheric radon observations at the Cape Point Station (CPT), in conjunction with associated meteorological measurements.

2. Methods and measurements

2.1. Site description

Various baseline atmospheric measurements are routinely performed at the coastal Cape Point Station, under the South African Weather Service (SAWS) management. The Cape Point Station is situated within a nature reserve about 60 km south of Cape Town at the southernmost tip of the Cape Peninsula (see Fig. 1). The station is part of the Global Atmospheric Watch (GAW) network of baseline stations, functioning under the auspices of the World Meteorological Organization (WMO). The station is located at an altitude of 230 m above sea level on a coastal cliff (S 34.353 211° and E 18.489 683°). A range of continuous trace-gas measurements have been performed at the Cape Point Station (including CO, CO_2, CH_4, O_3, N_2O, and halocarbons) for the past 30 years (Brunke et al., 2004). More recently atmospheric radon and gaseous elemental mercury (GEM) has also been added to the list of continuous measurements (Slemr et al., 2013).

2.2. Radon measurement instrumentation

Since 1999, the Australian Nuclear Science and Technology Organisation (ANSTO) designed detector have been continuously measuring hourly atmospheric radon concentration levels at the Cape Point Station. The metrology and performance features of the ANSTO-built two flow-loop, dual-filter detectors have been well characterised in previous publications (e.g. Whittlestone and Zahorowski, 1998; Brunke et al., 2004; Chambers et al., 2011; Griffiths et al., 2016). Incremental improvements in the detection sensitivity of the Cape Point Station radon instrument resulted in a lower limit of detection (LLD) that reduced from an initial 42 ± 13 mBq/m^3 down to 33 ± 10 mBq/m^3 by 2001 (Brunke et al., 2004). In 2011, an upgraded version of the radon detector was installed, incorporating numerous system design improvements including an automated calibration system and a delay volume designed to further reduce potential thoron in-air within the detection chamber. This detector upgrade resulted in further improvements to the accuracy and reliability of the radon-in-air activity concentration measurements at Cape Point, with a new LLD of 25 ± 8 mBq/m^3.

The radon detector's sample inlet is located 30 m above ground level on an aluminium mast overlooking the coastal cliff at the Cape Point Station. The current detection system has a radon activity concentration measurement resolution of 30 min, which is further processed to an average 1-h temporal resolution. Instrumental background counts (accumulation of ^{210}Pb on the detector's second air filter) are measured on a three-month basis. The standard deviation of these background activity measurements is equivalent to 5 mBq/m^3 (Brunke et al., 2004). The radon concentration uncertainty for hourly measurements, taking into consideration the standard deviation of monthly calibration estimates (~2%) and the ± 4% accuracy of the calibration source, is of the order of 15% at 100 mBq/m^3 and 9% at 1 000 mBq/m^3 (Chambers et al., 2014).

2.3. Wind measurement instrumentation

Meteorological measurements made on top of the aluminium mast