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Xenon and radon time series analysis: A new methodological approach for characterising the local scale effects at CTBT radionuclide network

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HIGHLIGHTS

- A new methodology of time series analysis has been tested on ^{133}Xe and ^{220}Rn.
- Characterisation of local effects at different CTBT/IMS monitoring stations.
- This study might help assess relative influence of near and far field air.

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ABSTRACT

A new methodology of time series analysis has been tested on ^{133}Xe and estimated ^{220}Rn activity concentrations in order to characterise the site response of four different CTBT/IMS monitoring stations. Seasonal variability of ^{133}Xe and ^{220}Rn at these IMS stations and the role played by different meteorological parameters on such variability have been quantified. As xenon and radon are both noble gases with similar physical characteristics but very different source terms, the methodology adopted in this comparative study, once coupled to analysis of radioxenon emission time series sampled at nearby NPPs or IPFs and to direct measurements of ^{220}Rn at IMS sites location, might help assess relative influence of near and far field air on IMS radioxenon detections. Possible applications of the adopted methodology to radioxenon categorisation schemes are also discussed.

1. Introduction

Comprehensive Nuclear-Test-Ban Treaty (CTBT) monitoring process relies on the capability of the International Monitoring System (IMS) to physically discriminate among different sources of emission of radioxenon, such as nuclear power plants (NPPs) and isotope production facilities (IPFs) (De Meutter, 2016). To reliably distinguish between legitimate and illegitimate (due to nuclear weapons tests) emission scenarios is a difficult task because of low radioxenon activity concentrations observed in the atmosphere after low-yield underground tests. Both low-release rates of radioxenon from such tests and its high atmospheric dilution make this task difficult. Therefore the minimum detectable concentration (MDC) of radioxenon in the atmosphere required by International Monitoring System (IMS) for monitoring stations has been set at $\leq 1\,\mathrm{mBq\,m^{-3}}$ (Auer, 2010).

In this regard, an important aspect of the CTBT monitoring strategy has been the development of categorisation schemes to help assess whether or not a measured activity concentration has to be considered anomalous, given an IMS station particular detection history. The categorisation scheme currently adopted by the CTBTO has three different levels while a scheme based on five levels, also involving analysis of isotopic ratio and possible source region estimation through Atmospheric Transport Modelling (ATM), has been proposed (Postelt, 2014).

Due to worldwide locations and heights of the IMS stations, meteorological patterns are expected to affect the measuring process in different ways. In this regard, radioxenon time series analysis carried out in Plastino (2010) suggest that outlier occurrence has been influenced by meteorological parameters, which are also measured at the IMS stations. Different emission scenarios are possible from legitimate sources, namely continuous or batch releases. At a downwind IMS station, these emission scenarios manifest themselves in the observational record either as unusual spikes of activity concentration or as fluctuations in the background activity concentrations (Saey, 2006). Besides monitoring Treaty-relevant radionuclides, IMS stations also detect naturally occurring radionuclides and their decay products. An example in this regard is ^{212}Pb ($T_{1/2} = 10.64$ h), a progeny of ^{220}Rn ($T_{1/2} = 55.6$ s), detected daily at the IMS stations. The term radioxenon refers to the following isotopes: ^{133}Xe ($T_{1/2} = 5.24$ d), ^{135}Xe ($T_{1/2} = 9.14$ h), ^{133m}Xe ($T_{1/2} = 2.19$ d) and ^{131m}Xe ($T_{1/2} = 11.84$ d), where

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also isomeric states are considered. Radioxenon is mainly of anthropogenic origin and its production, as opposed to radon and its progeny, can be expected not to be of local origin. It is instead advected to the IMS stations from neighbouring NPPs or IPFs localised in the far atmospheric field. Instead ^{220}Rn, being part of ^{232}Th decay chain, is a naturally occurring radionuclide which, having a short half-life, can be considered indicative of nearby soil conditions and radon turbulent transport near the exhalation site (Steinhäusler, 1996). Detailed description and modelling of how different local meteorological conditions and their seasonal variability can affect ^{220}Rn emission from soil, and consequently ^{212}Pb atmospheric dispersion, can be found in Baldacci (2010). Furthermore, xenon and radon, being both noble gases and hence chemically inert, are not expected to be removed from the atmosphere by processes of dry and wet deposition, which instead influence considerably radionuclide particulate.

The aim of this work is to characterise the variability of time series of two noble gases, namely ^{133}Xe and estimated ^{220}Rn, in order to investigate the site response of the IMS stations to signals with very different source regions but similar physical behaviour in the atmosphere. Hence, the purpose of the analysis is first to investigate the harmonic and residual contents through spectral analysis and then to evaluate cross-correlations with meteorological parameters. Results obtained from spectral analysis of the IMS time series, e.g. percentage of harmonic content, local Hurst exponent dynamic, and possible occurrence of frequency coupling in meteorological and radionuclide time series, could be employed as additional flags in station specific categorisation schemes. In order to do so, the proposed methodology should necessarily be coupled with analysis of regional emitters emission time series and with forward simulations with ATMs. Time resolution employed should depend in this case on the relative distance between a given emitter and the selected IMS station location, as described in De Meutter (2018). This way, the methodology adopted in this paper could give quantitative insights on the relative influence of local meteorological conditions on radioxenon outlier occurrence at IMS stations.

2. Data analysis

In order to characterise the site response of the selected IMS stations in terms of time series variability, activity concentrations of ^{133}Xe were compared with ^{220}Rn ones, estimated from its progeny ^{212}Pb, and with the following meteorological parameters: relative humidity RH(%), temperature T(°C), atmospheric pressure P(hPa), wind direction α - wind direction is measured in degrees clockwise from North - and wind speed v(m s^{-1}). These parameters are measured at each IMS station location.

All time series data were analysed using a software for time series analysis which allows to determine the harmonic content even in case of missing data, making use of the Generalised Lomb-Scargle Periodogram (Lomb, 1976), to perform detrended cross-correlation analysis (Podobnik and Stanley, 2008) of residual time series and to estimate its Local Hurst exponent H_t, obtaining quantitative information on long-term correlations (Ihlen, 2012). A detailed mathematical description of the software used for data analysis, along with its applications to other radionuclide time series, can be found in Bianchi et al. (2018a, 2018b), Bianchi and Plastino (2018). Based on radioxenon data availability, four IMS stations were chosen for the analysis and then results were compared with estimates of ^{220}Rn activity concentrations and meteorological parameters. The four stations ID, their sampling time, and the investigated periods are the following:

- USX75, Charlottesville-USA (12 h), 7/6/2011–15/3/2015;
- SEX63, Stockholm-Sweden (12 h), 1/9/2012–31/3/2016;
- DEX33, Schauinsland-Germany (24 h), 30/6/2013–31/3/2016;
- CAX17, St. John's-Canada (24 h), 14/8/2014–31/3/2016.

Differently from radionuclide data, meteorological parameters have higher sampling time and hence daily averages have been calculated before carrying out comparative spectral analysis. Consequently, information about variability at time scales shorter than two days can not be extracted in this case. Furthermore, stations SEX63 and USX75 have 12 h sampling time and data from those stations have also been averaged to match the 24 h sampling time of DEX33 and CAX17 stations. For SEX63 station, wind speed time series contained too many missing data during the available period and was hence excluded from the analysis.

Following Werzi (2010), ^{220}Rn source term was estimated from its progeny ^{212}Pb activity concentration, which is measured daily by the radionuclide (RN) component of the IMS, making use of the theoretical expression

$$C_{^{212}Pb} = \frac{\lambda_{^{212}Pb}}{\lambda_{^{220}Rn} - \lambda_{^{212}Pb}} C_{^{220}Rn} = 0.0015 \quad C_{^{220}Rn}$$

having introduced the following decay constants: $\lambda_{^{212}Pb} = 1.81 \cdot 10^{-5} s^{-1}$, $\lambda_{^{220}Rn} = 1.2 \cdot 10^{-2} s^{-1}$. A detailed modelling and comparative study of ^{220}Rn and ^{220}Rn progeny concentration, based on measurements performed indoor, can be found in Guo et al. (1995).

3. Results and discussions

In the following section, some results obtained from the spectral analysis are presented. Fig. 1 shows the analysed ^{133}Xe and ^{220}Rn data, with a sampling time of 24 h. For USX75, a very high outlier of xenon ($> 30\sigma$), occurring on November 2011, was removed before performing spectral analysis. Table 1 summarises harmonic and residual percentage, along with the carrier signal and its percentage, obtained from the analysis both for radionuclide and meteorological parameters time series.

As explained in Bianchi et al. (2018a), a periodic component is identified when the corresponding peak in the generalised Lomb-Scargle spectrum is above a given threshold, chosen in order to have a 95% confidence level of discriminating harmonic components from background noise. Periodic components are then filtered using a notch filter and their percentage weight on the spectrum is evaluated. Once all the periodic components are filtered out, residual time series and their characteristic behaviour can be investigated. Fig. 2 shows normalised residual time series of ^{133}Xe and ^{220}Rn for the four IMS stations considered. In Fig. 3, normalised residuals obtained both from radionuclides and meteorological parameters time series are shown for station USX75 only. More detailed comparison of ^{133}Xe and atmospheric pressure residuals at this station can be found in Fig. 4, in which anticorrelation among the two variables is clear.

In Fig. 5, local Hurst exponents H_t obtained for ^{133}Xe and ^{220}Rn residual time series, as a function of windows of small length, are compared for the four stations. Different values of the local Hurst exponent identify how the characteristics of residual time series change with time. As described in Bianchi et al. (2018a) values of the local Hurst exponent ranging between 0 and 0.5 are indicative of anti-correlation in the residuals, $H_t = 0.5$ corresponds to white noise while when $0.5 < H_t < 1$ residuals are characterised by long-range correlations. $H_t = 1$ corresponds to pink noise and $1 < H_t < 1.5$ indicates stronger long-range correlations. A value of $H_t = 1.5$ corresponds to Brownian noise. Fig. 5 shows that local Hurst exponents oscillate around their average value. Regarding station SEX63 (panel c,d), ^{133}Xe and ^{220}Rn local Hurst exponents seem to do so in an anticorrelated fashion. To give a physical interpretation of such behaviour further investigation is needed, possibly employing data collected at higher sampling rates. In Fig. 6, yearly averaged values of local Hurst exponent H_{μ_t}, obtained for meteorological parameters, are summarised for station SEX63. In this case, external values are not representative of the whole year, but of the available data in that portion of the considered year. Table 2 summarises the average values of local Hurst exponents H_{μ_t} obtained from the analysis of the four IMS stations.