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RADON, AS A TRACER FOR MIXING HEIGHT DYNAMICS - AN OVERVIEW AND RADO PERSPECTIVES*

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*Abstract***.** This paper analyses the various existing experiments using the natural isotope Radon – 222 in estimating the planetary boundary layer (PBL) dynamics and particularly the regional pollutants mixing height. The paper will propose an instrumental set-up in frame of RADO project (Romanian 3D Atmospheric Observatory) based on meteorological tower instrumentation (standard meteo station with 30 and 60 m measuring levels) supplemented by eddy covariance for extensive studies and using well established relationships for assessing mixing height from surface measurement. This will include also daytime comparisons with MAP3D (Mesoscale Air Pollution 3D model) simulations in Bucharest-Magurele station area. Finally LIDAR observations for PBL height validation will be used for development of local Radon tracing function. Further potential development of RADO will be argued.

Key words: Radon, surface emission, atmospheric tracer, planetary boundary layer.

1. INTRODUCTION

"Horia Hulubei" National Institute for Physics and Nuclear Engineering has a large environmental survey program covering radioactivity in air, soil, water, crops, as well as its own meteorological observations. The meteorological observations are used for assessment of radionuclide atmospheric transport modelling. Current upgrades under national and international research financing offer new possibilities to link nuclear and environmental studies with atmospheric science. Radon is monitored biweekly for the purpose of normal survey during the working hours, but it will be continuously monitored in the surface air. The new

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equipment planned to be on site next year, including eddy covariance, lidars, and ceilometers will provide direct measurements of turbulence, sensible and latent heat flux, cloud cover and mixing height. This opens the possibility to study Radon as an atmospheric tracer and the present paper revises the topic and considers an instrumental setup in the frame of RADO project (Romanian 3D Atmospheric Observatory).

2. RADON AND ATMOSPHERIC SCIENCE

The diurnal structure of the atmospheric boundary layer (ABL) has an important impact on the dispersion of chemical compounds [1]. The main characteristic of the ABL is its turbulent nature that drives the scalar transport with a broad range of spatial and temporal scales. Turbulent eddy motions transport and mix primary and secondary pollutants throughout the ABL. Large-scale turbulent eddy motions (e.g. thermals and subsidence motions) characterize the daytime convective boundary layer, while the nocturnal boundary layer has significantly smaller eddies. In large-eddy simulation (LES), the largest eddies that are responsible for the turbulent transport of the scalars and momentum are explicitly solved, whereas the smallest ones that are mainly dissipative are parameterized using a sub grid-scale (SGS) model.

A particular attention was recently paid to the nocturnal stable layer, for which the diagnostic and prognostic modelling represents a major challenge for atmospheric physicists and meteorologists. Until this issue will be adequately addressed, our ability to forecast future climate trends with reasonable confidence will be compromised by the large differences observed between measured and modelled near surface climate parameters, and indeed, even between those of comparable models – particularly for high latitude regions.

The ABL height also referred as mixing height (MH) determines the vertical mixing of the substances in the boundary layer due to the turbulent exchange and in this way, defines the distribution of pollutants. Vertical diffusion depends mainly on the presence of an inversion layer during the night and the thickness of the mixing layer during the day.

During the last decades, many studies about indoor and outdoor radon concentration were performed, mainly for epidemiological purposes, and this will be not covered in the present paper.

The description of the atmospheric boundary layer on the base of measurements of the natural radioactivity is one of the methods used in the evaluation of mixing processes in the atmosphere [2]. Observations of atmospheric ^{222}Rn are very useful in the evaluation of climate models for simulating transport, transformation and removal processes of gases and aerosols [3]. Regional transport models use also radon for validation [4].

The World Meteorological Organization (WMO) established the Global Atmosphere Watch (GAW) Program to investigate the role of atmospheric chemistry in global climate change based on data analysis for many stations. When the results given by these stations are analysed, it is important first, to consider how seasonally changing fetch regions and local mixing affect long-term observations at each site, and second, how the observations are comparable between stations in the network in a global context. To these extends, hourly ^{222}Rn concentration data are used in conjunction with other observations [5].

^{222}Rn is a naturally occurring, radioactive, noble gas, with a low solubility in water. It is emitted from terrestrial surfaces at an approximately constant rate on diurnal timescales and the emissions are assumed to be uniform on local to regional scales. The half-life of ^{222}Rn (3.8 days) is optimum for boundary layer mixing studies, because it is long compared with typical turbulent timescales (less than 1 hour), but short enough to constrain its concentration in the free troposphere (typically 1-3 orders of magnitude lower than the near surface values). The combination of these properties makes radon an excellent tracer for vertical mixing studies in the boundary layer. As a radioactive species, its decay chain is: ^{222}Rn => ^{218}Po => ^{214}Pb => ^{214}Bi => ^{210}Pb. The decay constants of ^{222}Rn, ^{218}Po, ^{214}Pb and ^{214}Bi are equal to: 2.11×10^{-6}, 3.80×10^{-3}, 4.31×10^{-4}, and 5.80×10^{-4} s^{-1}, respectively. Note that we consider a direct transformation of ^{214}Bi into ^{210}Pb because the half-life of ^{214}Po (daughter of ^{214}Bi) is very short (164 μs). We also consider that ^{210}Pb has a half-life of 22.3 years, being an inert scalar in respect to the temporal scales considered here.

The techniques such as acoustic measurement, radio analysis and analysis of the mechanical turbulence with sonic anemometer, could furnish many information on ABL processes but their use is limited by their costs and complexity. The technique based on the concentration measurements of the radon gas is simple and very useful to describe the temporal evolution of the pollutants in the atmosphere.

Since the late 1970s, researchers pointed out the relevance of radon detections for the characterization of the boundary layer properties [6, 7]. ^{222}Rn concentration has an inverse linear dependence on the mixing height, with an unexpectedly high linear correlation coefficient of about −0.96 and conversely, it is much less dependent on wind speed than it was expected. A relationship between variations in ground level radon concentration and the atmospheric state of stability or turbulence has been reported several times, but models for obtaining quantitative data on the height of the mixing layer are still studied [8]. An example of the correlation between radon concentration and mixing height is given in Figure 1 [9]. The correlation, even it is not very high, it is in any case significant (–0.4). Note that the mixing height was not directly measured, but it was obtained as an output of CALMET, a meteorological pre-processor based on profile of wind and temperature measured two times per day and some approximations.

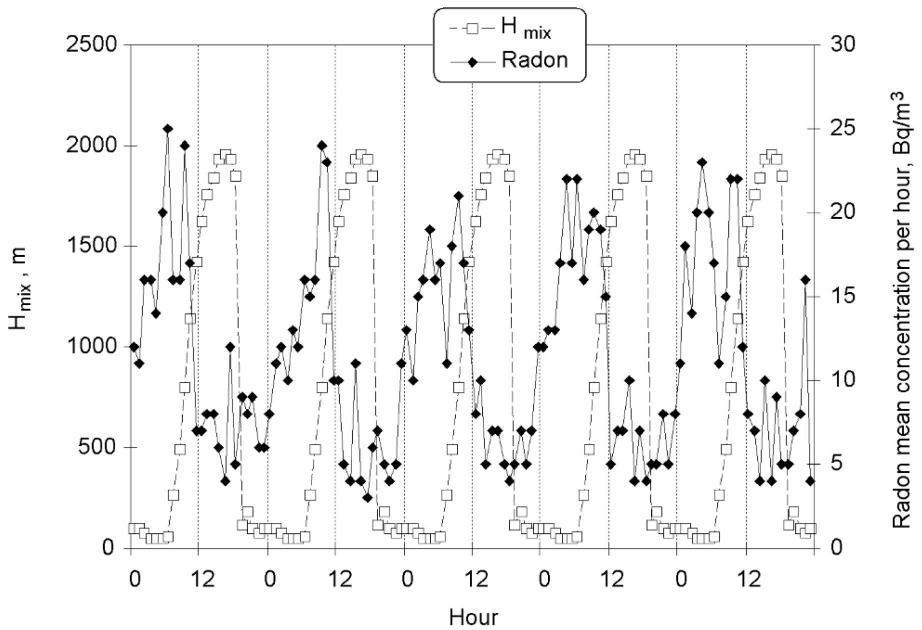

Fig. 1 – Correlation between radon concentration and mixing height [9].

Many previous attempts which correlate the MH with radon concentration, consider the emanation quite constant in time and space, a gross approximation [10]. First, only part of the produced ^{222}Rn emanates into air filled pore space from where it might escape into the atmosphere and the fraction emanating may depend on grain size. Second, the differences in grain size and soil moisture modulate the gas diffusivity and thus the fraction of emanated ^{222}Rn, that may reach the atmosphere before decay. Thus, the proportion of ^{222}Rn produced that escapes into the atmosphere is variable and depends on factors others than ^{226}Ra content. The correlation between ^{222}Rn and terrestrial gamma dose rate (GDR) showed that both parameters are similarly affected by the radionuclide content in soil and soil moisture. Most of the spatial variation in ^{222}Rn flux may be explained by the variation in radionuclide activity in soils derived from different parent material. Soil moisture has been shown to have similar effects on ^{222}Rn flux as it has on GDR, excepting for short time periods during precipitation events.

For a proper interpretation of the link between radon concentration and mixing height, it is mandatory to analyze the correlation between radon concentration and other meteorological variables. The correlation between radon and temperature is very important [9]. A frequently encountered daily pattern is characterized by the accumulation of radon during the night time, and until the early hours in the morning, followed by a decrease. In fact, the highest radon concentration is recorded in the first hours in the morning, when the temperature is low; on contrary, the lowest values are recorded in the first hours in the afternoon, when the temperature is high. During warm months, the radon concentration shows a well-defined and modulated temporal pattern, all days are similar: nocturnal stability and convective mixing during the daytime. This pattern was observed typically on sunny days, when the sky is clear both during the daytime and night time and there is little ventilation. During the cold months, the high-pressure periods are sporadic and the advection often occurs. The diurnal mixing is weak and of limited duration. Furthermore, more recent results [11] show that radon and radon daughters concentrations are approximately homogeneous with altitude in the nocturnal stable layer and that they undergo a rapid transition to background values above the mixing height (in the so called residual layer) during the daytime.

Considering the time variability of radon concentration in air, wavelet analysis was used to better identify the features between radon and atmospheric parameters [12]. For one year period, the correlations have been analysed and the correlation coefficients were found for:

– wind speed −0.457;
– temperature −0.477;
– relative humidity –0.634;
– precipitation −0.061;
– pressure 0.258.

A sudden increase of concentration was observed due to the collapsing of the daytime ABL. This phase lasts between 6 to 8 h; the sharp collapsing of concentration due to the rapid growth of the ABL is between 09:00 a.m. and 02:00 p.m.; a remarkable feature of radon atmospheric phenomenology is the response time of radon exhalation and therefore, the concentration at the start and at the end of the precipitation event. In particular, it was found that even a short term intense precipitation event can have a prolonged effect on the concentration in the atmosphere due to water saturation of the first layers of soil. The sensitivity of radon exhalation and concentration to small amounts of precipitation, points clearly towards the need of a parameterization of the process within global models. The availability of micro meteorological measurements in combination with radon concentration and flux data would allow an even deeper investigation of radon dispersion and boundary layer properties. Future investigation will be dedicated to determine the effect of precipitation intensity on the radon surface flux and surface layer concentration.

From the modelling point of view, the main limitation in simulating the diurnal ABL cycle resides in the difficulty of resolving both the small scales that characterize the nocturnal boundary layer and the large ones, of the daytime case, with the same sub-grid scale model. Modern models have been recently used extending the study of the control exerted by turbulence on ^{222}Rn and its progeny atmospheric dispersion. A study for convective conditions concludes that the turbulent properties of the atmospheric convective boundary layers are important to study the dispersion and the transport of the radon family [13]. An extension to the full range of stabilities characterizing atmospheric boundary layers was also done [14]. The simulation starts with radiosonde data, analyzing the wind and temperature field and supposing a typical exhalation rate of the parent (the source term *S*0), and secular equilibrium of daughters in the soil. It could be expected that the species produced (decaying) faster than the others may appear (disappear) in (from) specific regions of the ABL due to the turbulent transport and mixing process. The timescale of the turbulent transport and how it relates to the timescale of the radioactive decay is one of the governing parameters of the process. The deviation from the secular equilibrium is often used as an indicator of the residence time in the atmosphere or as an indicator of the atmospheric stability. Around sunset, the vertical motions are suppressed due to the cooling at the surface. This cooling results in a temperature stable stratification and in the formation of a thin boundary layer isolating the surface from the upper atmosphere layer. In this layer, turbulence decays, leaving a residual layer instead of the well-mixed convective the layer. The nocturnal BL is characterized by very high concentrations and important concentration vertical gradients. During the night time, *S*0 is constantly emitted and, due to the stability of the NBL, it is accumulated close to the surface. *S*0 and its short-lived daughters undergo the unidirectional chain of reactions with the radon concentration flowing into the daughter's. As a result, *S*0 progeny also accumulates close to the surface. At sunrise, the solar heating causes thermal plumes to rise. These plumes expand up to the top of the atmospheric boundary layer where a thermodynamic equilibrium is reached. During the morning transition and the development of the unstable boundary layer concentration of all radionuclei abruptly reduces in spite of the fresh emission of *S*0. The time evolution of the concentrations is the result of the combined effect of the divergence of the fluxes and the radioactive decay contribution, as well the disequilibrium (non secular) is more important under stable stratification in the nocturnal boundary layer.

The assumption of a constant emanation rate on the area of interest is restrictive and depends on measurement height (fetch must be homogeneous for about 100 times of the measurement height). At higher level of measurement the pattern of radon concentration is influenced by the non-homogeneity of ”foot print” depending on wind direction, wind velocity and shows distinct diurnal and seasonal variability. For example, at Cabauw, The Netherlands [15], for 20 m height, under low wind speed conditions, and typical summer conditions for that region, the local contributions are predominant in the diurnal radon measurements. When radon concentrations are analysed in correlation with wind speed (lower than 3 m s^{-1} and higher than 7 m s^{-1}), it is observed a distinction between these two wind regimes. The low wind speed signal exhibited a pronounced diurnal cycle with large annual average and large nocturnal gradients. In contrast, the high wind speed signal showed a little diurnal cycle. Under high wind speed conditions, the nocturnal boundary layer is weakly stratified or absent, and radon is mixed through a deep column leading to a weak diurnal signal.

The above results show that we have a quite complex dependence of radon concentration and mixing height and there are many local influences which must be detected. For sunny days, it was obtained a correlation [16] between measured mixing height at time t and ^{218}Po concentration delayed with 2 hours, with correlation coefficients depending on site.

3. OUTDOOR RADON MEASUREMENT

The ^{222}Rn concentration can be estimated from its daughter short lived beta radionuclides taking into account the following assumptions:

a) the daughter radionuclides are in equilibrium one to the other and to ^{222}Rn (this is not usually true, but the isotopic ratio in open air is estimated to be near 1 (about 0.8–1.1);

b) the detector efficiency for counting ^{214}Bi and ^{214}Pb is similar;

c) the concentration of ^{220}Rn daughters in the surface air is one order of magnitude lower than those of ^{222}Rn.

The accuracy in the evaluation of the radon concentration based on short lived beta measurements is estimated to be 20%. Usually, the aerosol samples are collected on filters and air volume is measured by a flow meter. After collecting, the filters are measured at different time intervals (5, 60 minutes, 360 minutes) [17] or are measured in alpha/beta counters at 3 minutes, 5 hours and 5 days, respectively [18]. This method is time consuming and not appropriate for continuous monitoring, but it is used in normal environmental survey. For example, in Romania, it is the basic method for our national radioactivity survey network, as well as for routine survey in IFIN-HH. For a 5 hours sampling time, results from IFIN-HH are given in Table 1, showing a minimum level of 0.4 Bq m^{-3}. Depending on microclimatic and geological conditions, outdoor radon average concentrations in different regions of Romania are from 1,200 mBq m^{-3} to 13,065 mBq m^{-3}.

Table 1

Aerosols Global Alpha, 3 minutes after the end of aspiration

Filter $P_{i\,(I=1...8)}$	Aspiration time	Activity [Bq m^{-3}]
1	08^{24}-14^{24} (11.06.05)	1.331±0.027
2	14^{24}-19^{24} (11.06.09)	1.252±0.03
3	19^{24}-24^{24} (11.06.09)	1.098±0.029
4	24^{24}-05^{24} (12.06.09)	3.559±0.054
5	05^{24}-10^{24} (12.06.09)	3.70±0.06
6	10^{40}-15^{40} (12.06.09)	1.397±0.033
7	15^{45}-20^{45} (12.06.09)	0.411±0.018
8	20^{45}-05^{45} (12.06.09)	1.51±0.035

The continuous measurement of radon concentration in air near the surface can be performed by the alpha-counter “AlphaGUARD”, a pulse-counting ionization chamber [9]. This radon monitor is suitable for continuous monitoring of radon concentrations between 2–2,000,000 Bq m^{-3}. The lower limit excludes the usage in our case. Semiconductors detectors are also used in various configurations with better performance when it is used alpha spectrometry. Immediately after decaying of radon, about 80% – 82% of ^{218}Po occurs as positive ions that are attached to air gas molecules and water particles within 10^{-7} seconds [16]. These formations are called clusters (free atoms and clusters are called unattached fraction). Then the clusters can generate bigger particles called attached fraction. During measurements, the air together with all particles is drawn through a filter with adjusted flow rate. A special semiconductor, PIPS CAM detector, is placed above this filter and the alpha radiation emitted by polonium, which is separated outside of air stream and deposited on filter, can be detected using this PIPS CAM detector [16].

Dual Si detectors are also used for alpha and beta spectrometry analysing a moving filter with gamma compensation (BAB-A6 monitor from Saphymo Ltd.).

Non commercial instrumentations have also high performance. For example [19], radon concentration measurement is based on alpha spectrometric measurement of ^{218}Po which is collected electrostatically on a passivated implanted planar silicon (PIPS) detector. The sensitive volume is divided into two regions by a metal wire-screen and the PIPS detector. The lower region of the instrument is a glass sphere which is covered internally with silver, while the upper region is a glass cylinder. The wire screen and the silver surface are electrically connected and the PIPS detector is isolated from them. A potential of 8 kV is applied between the detector and the silver screen surface generating an electrostatic field that moves the charged ^{218}Po to the detector surface. For one hour measurement a detection limit of 0.17 Bq m^{-3} is assured.

Dual-flow loops, two-filter detectors, developed at ANSTO [20], were used at each of the sites. The principle of operation of these detectors is illustrated in Figure 2. The external flow loop draws sample air through the detector at a low flow rate. Between the inlet and the first filter the sample is delayed to reduce ^{220}Rn (Thoron) content to negligible levels. The first filter removes aerosols and ambient radon progeny. The air then enters the radon delay volume where it is circulated at a high flow rate by the internal flow loop, passing repeatedly through the second filter. A fraction of the new radon progeny generated in the radon delay volume is captured on the second filter. These short-lived, alpha emitting radon progeny are counted using an assembly consisting of a ZnS scintillator, photomultiplier tube and counting electronics. The performance is 30–40 mBq m^{-3}, the lower limit of detection. At this performance, with two instruments at 2 and 50 m, the information on low level temperature inversion on radon profile can be obtained. Close to the surface, radon concentrations build up at night with the formation of the nocturnal stable boundary layer (SBL). In some nights, the radon data indicates that the local SBL height is below 50 m and the surface is thermodynamically decoupled from the air aloft.

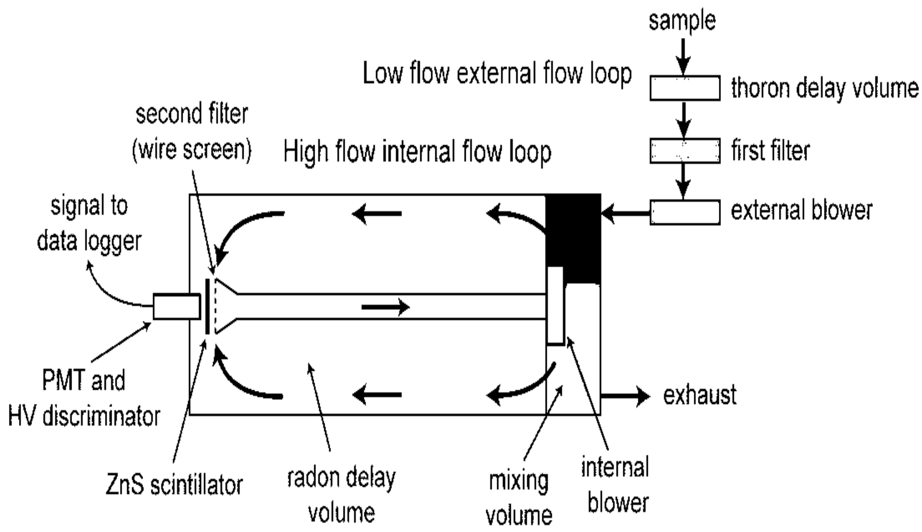

Fig. 2 – Principal of operation for dual-flow loops two-filter detectors [20].

Considering preliminary data in IFIN-HH and the need to measure at a height over 20 m, it seems that the instrument must have a better performance than 0.1 Bq·m^{-3} for one hour sampling. The choice of the best acquisition will depend on cost, maintenance and calibration needs. Note that IFIN-HH will produce the national radon standard, but it still has not a calibration chamber.

4. PROPOSED APPROACH FOR RADON CORRELATION WITH MIXING HEIGHT IN THE FRAME OF RADO

The characteristic of IFIN-HH site and the Magurele area is well know in respect of actinide concentration in soil, external gamma dose rate, land use and roughness. There is no rationale for a radon emanation varying with more than a factor 2 and the phosphogypsum was not used for buildings. This can be further checked for far area, farther than 3 km. Land use and roughness non homogeneity (Fig. 3) affect the wind profile and make inapplicable the classical procedures to assess the atmospheric stability.

Fig. 3 – Google map of area around IFIN.

Recently, it was developed a methodology based on a mixture of measurements (temperature gradient and wind direction standard deviation at 30 and 60 m) and recent results about air flow over complex terrain have been obtained. Eddy covariance will be soon used delivering accurate information on local turbulence, fluxes, local Monin-Obukhov length and a robust assessment of stability in the surface layer. Remote sensing will be used for the profile of temperature, humidity and wind. Ceilometers will provide data on cloud base and top, while lidars will provide PBL height and substructures, and layers in the free troposphere. With accurate radon measurements we will be able to have a full data base on the same site (this was not possible in the majority of past studies). Variability of emanation rate with soil water and temperature, as well as basic processes in atmospheric transport, have been analyzed in the past by Romanian researchers, so they will be easy to incorporate in more modern models. Because the wind rose (Fig. 4) indicates a potential influence from Bucharest town, radon measurement from the network of Health Ministry will be used in order to detect the local emanation.

Atmospheric species of terrestrial origin, including aerosols and climatically sensitive gases, are usually monitored at ground stations where air masses are sampled and their composition determined. Consequently the composition will depend upon the stations' footprint. A station's footprint is the region most likely to contribute to the atmospheric composition of air masses arriving at the station (Fig. 5). In case of radon, we expect some influences of Bucharest town and the data will be analyzed in correlation with wind direction and speed. Consequently, we can analyze the correlation with measured mixing height extracting the other influences. Because the full equipment will be in operation in spring 2011, the results are not expected before 2012.

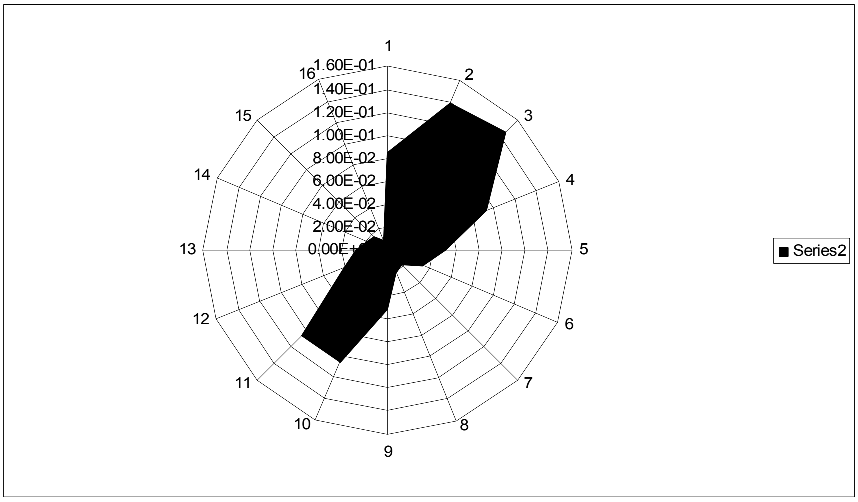

Fig. 4 – Wind rose.

The planed research will complete and explain the past observation of external dose profile. The increase of external gamma dose at 10 m was observed in correlation with strong temperature inversions (at 10 m, and 60 m) [21].

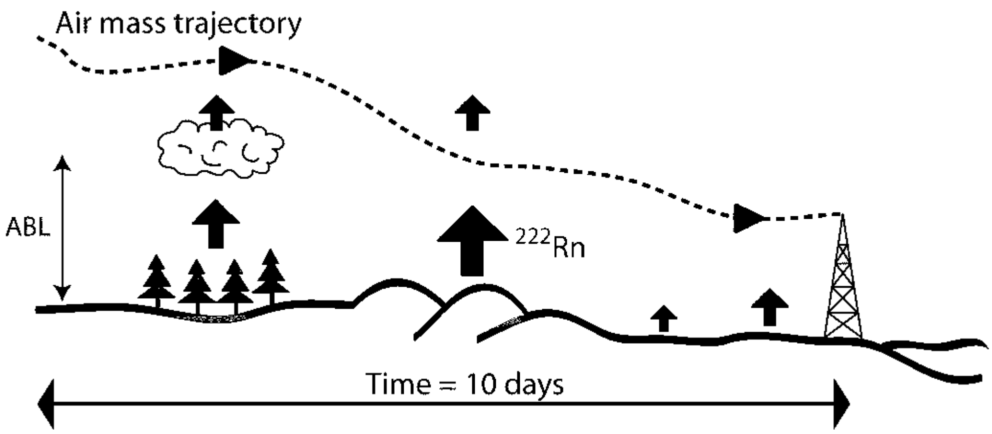

Fig. 5 – Schematic view of an air parcel's movement en route to a ground station.

5. CONCLUSIONS

The paper summarizes the actual status of research considering radon as an atmospheric tracer. The aim of this paper is to define a strategy to improve the knowledge related to the link between dynamics of outdoor radon concentration and mixing height. Due to past experience of IFIN-HH and considering the environmental radioactivity and the instrumentation developments in RADO project, there are opportunities to contribute to the topic and to expand the link between nuclear science, nuclear safety and atmospheric research. The impact of some meteorological factors such as wind direction, wind velocity, humidity, and temperature on short-lived beta radionuclides showed no simple statistical relationship [22]. The overall analysis suggests that longer data records are required in order to evaluate typical behavior, mean concentrations and trends of the outdoor short-lived radon progeny and radon itself.

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