

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

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# Mission


IFIN-HH is a public research organization dedicated to research and development in physical and natural sciences, mainly Nuclear Physics and Nuclear Engineering, and in related areas including Astrophysics and Particle Physics, Field Theory, Mathematical and Computational Physics, Atomic Physics and Physics of Condensed Matter, Life and Environmental Physics. In all these fields, IFIN-HH conducts theoretical and experimental research. Featuring a variety of nation-wide- scoped facilities including a Tandem Van de Graaff accelerator, a Cyclotron, a Multipurpose Irradiation Facility, a Radioactive Waste Processing Plant, the institute is an important part of the Romanian research infrastructure.

### Large Scale Facilities

- U120 Cyclotron
- FN-15 Tandem Accelerator
- Radioisotope Production Center
- Nuclear Waste Processing Center
- National Radioactive Waste Repository
- IRASM - Multipurpose Irradiation Facility
- VVRS Reactor - in decommissioning




**IRASM**



**Radioisotope Production Center**



**CYCLOTRON**



**TANDEM**

### Basic & Applied Research

- **Theoretical Physics:** nuclei, fields & particles, condensed matter, nonlinear phenomena, mathematical & computational physics;
- **Nuclear Physics:** structure & reaction mechanisms, hadronic matter, ion beam analysis, neutron diffraction, nuclear & atomic data, nuclear astrophysics, advanced detection systems;
- **Particle Physics:** calorimetry, Monte Carlo simulations, object oriented software, statistical data analysis, phenomenology;
- **Life & Environmental Physics:** low-dose radiobiology, nuclear risk, geophysical migration, control of soil erosion, dosimetry and radiation metrology.

### Production & Services

- Radioisotopes & Radiopharmaceuticals
- Technological Irradiations
- Radioactive Waste Treatment & Storage
- Decommissioning of Nuclear Instalations

### Other Activities

- Nuclear & Vacuum Engineering
- Informatics & Communications
- Training in Nuclear Activities

### International Collaborations

With international institutions:

- CERN (Geneva, Switzerland)
- EU (Brussels, Belgium)
- IAEA (Vienna, Austria)
- ICTP (Trieste, Italy)
- JRC (Ispra, Italy)
- INFN (Italy)
- GSI (Darmstadt, Germany)
- IN2P3 (Paris, France)
- GANIL (Caen, France)

With universities and research centers from abroad:

- Europe - 50
- USA & Canada - 11
- Asia - 3

Member of:

- JINR (Dubna, Russia)
- FAIR (Darmstadt, Germany)

Participation in large scale international experiments and projects

ATLAS, ALICE, LHC-b, DIRAC, FOPI, LCG, GASP, KASCADE, SPIRAL2

RODOS, EURONS, EURISOL

Around 250 publications / year in international journals

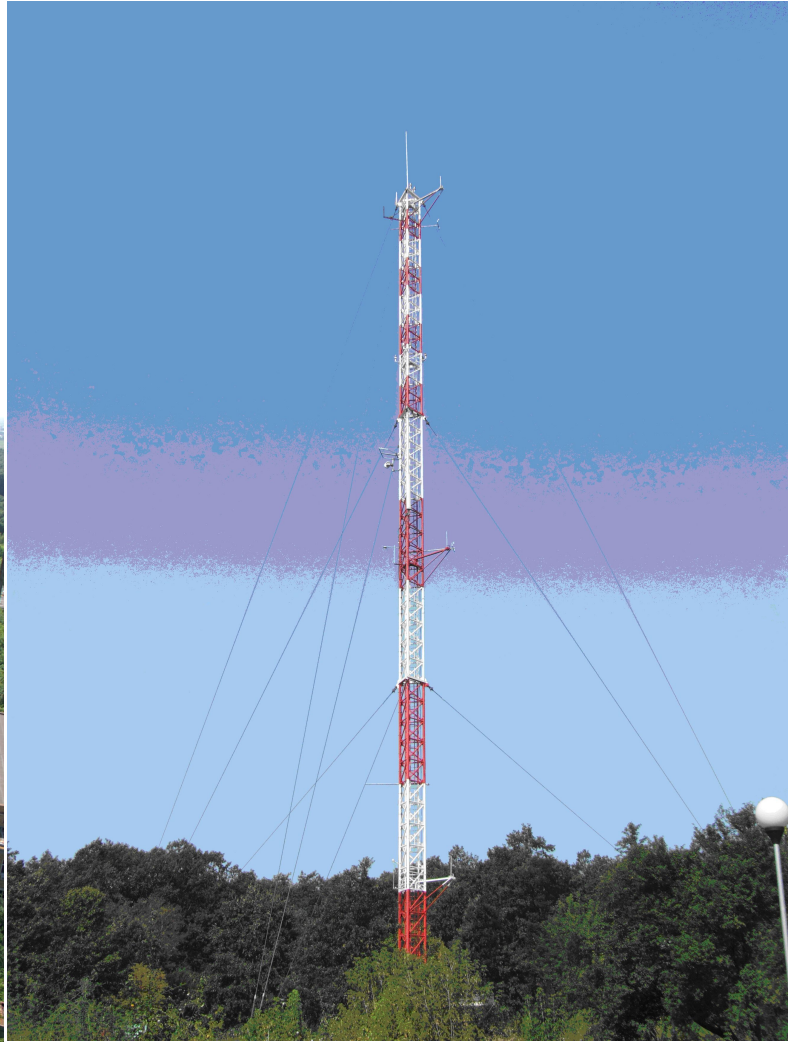
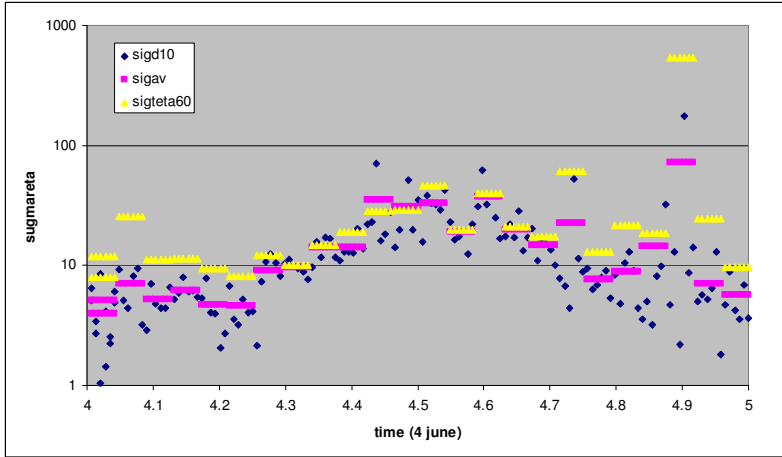
### Excellence in Research ...

- ... for Nuclear Impact on People, Environment and Material Goods
- ... on Nuclear Techniques and Methods for Material Analysis, Description and Certification
- ... on Nuclear Structure and Application
- ... on Materials and Biomaterials
- ... on Nuclear Interaction and Hadronic Matter

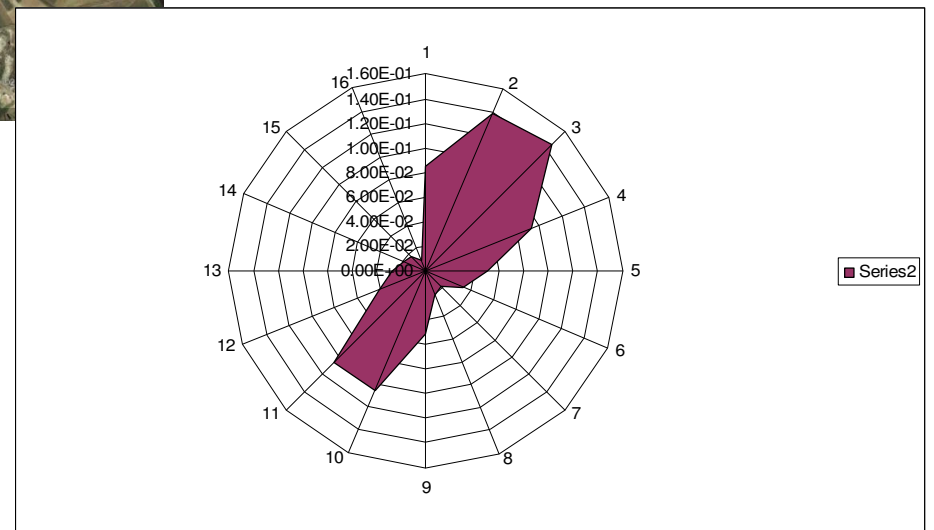
Computing facilities

Library

While focusing its mission on advanced investigations in atomic and sub-atomic Physics, IFIN-HH is also committed to widening the positive impact of the Nuclear on industry, other business areas, as well as on the society at large, via a diversified offer of unique professional services. Various applications of nuclear physics and spin-offs of basic nuclear research enable the institute to not only play an active promoter of domestic progress and modernity, but also bring a significant contribution to the public acceptance of the Nuclear. In tune with the mainstream topical research, IFIN-HH is asserting itself as a valid partner, in the realm of the Euro-Atlantic science and technology endeavor.



# Non homogen site



# Previous experience

Nuclear physics and nuclear accidents > atmospheric transport  
Chernobyl in Romania and IAEA

The integrated and comprehensive real-time on-line decision support system (RODOS) for nuclear emergencies in Europe was not designed initially to include the special radionuclide tritium.

RODOS customization and model development (tritium), incl. ATM

Remote sensing was too expensive.\?!

Second after Germany to operate RODOS in real time with own Meteorological Tower

Environmental Modelling for RAdiation Safety IAEA

General aim of programme

To improve capabilities in the field of environmental radiation dose assessment

(acquisition of improved data for model testing)

Large uncertainty due to atmospheric transport modeling

# RADO and IFIN

- The RADO project is under environmental section with main goal **REDUCE AIR POLLUTION**
- IFIN-HH is doing research in Nuclear Physics and associated fields
- Consequently the role of IFIN-HH in RADO is to make the link between air pollution and nuclear safety issues (atmospheric transport of radioactive pollutants)
- IFIN-HH has a meteorological tower, useful for the project. This is upgraded at the level of final goals of the project - robust, on line-real time atmospheric observations and related quantities needed
- Atmospheric observation must be upgraded by fast response sensors for wind, water vapour and CO<sub>2</sub>- eddy covariance method. This will be implemented first time in Romania providing data for turbulence, friction velocity, sensible and latent heat flux, and Monin-Obukhov length needed for modern atmospheric transport models. Non- homogeny of terrain around will be a challenge
- **As suggested by the consortium, we will implement a continuous survey of radon in the lower atmosphere - one of the tracer on atmospheric processes**
- Instrumentation will be completed with a ceilometer for cloud cover and mixing height assessment
- A local data centre will be linked with the RADO Data and Science Centre

# Influence of roughness change- generation of internal layer

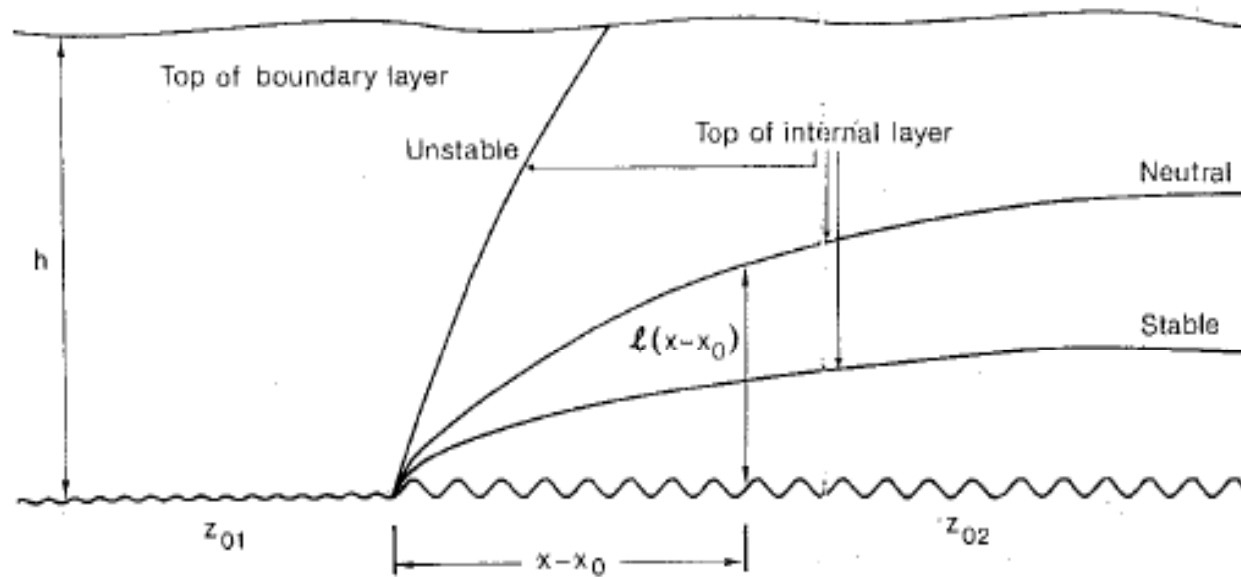
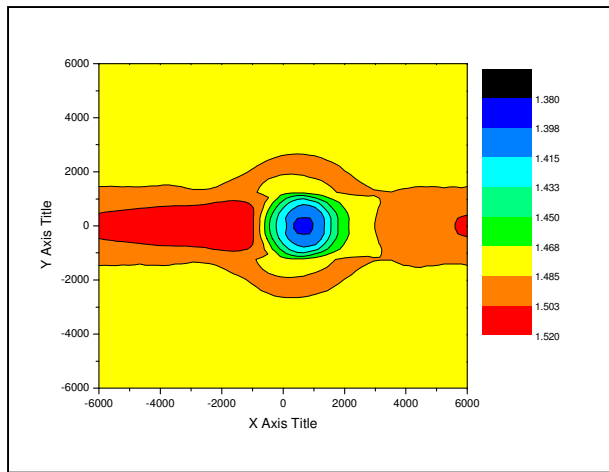


Fig.2.17a Variation of velocity internal layer formed at a change of roughness with stability.

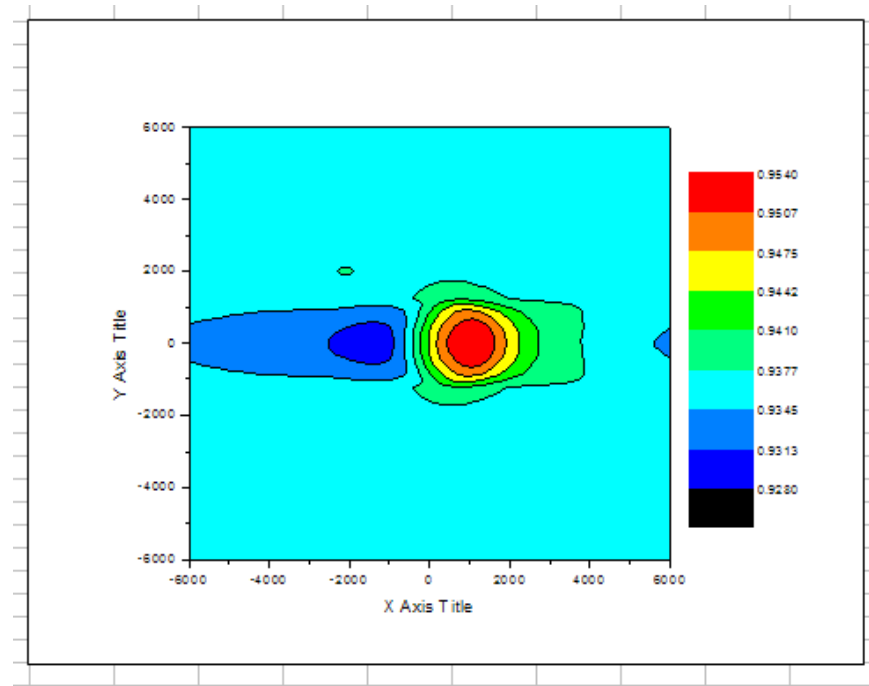
Profiles are affected by stability and roughness around. No classical procedure for assessing atmospheric stability is possible  
Most nuclear sites are not in open flat terrain

# FLOWSTAR



Wind field at ~60 m,  
Unstable atmosphere

Turbulence field at ~60 m,  
Unstable atmosphere



Combine temperature gradient, wind speed, wind direction, solar radiation and Standard deviation of lateral wind direction- 10 minutes time series



# Hourly standard report

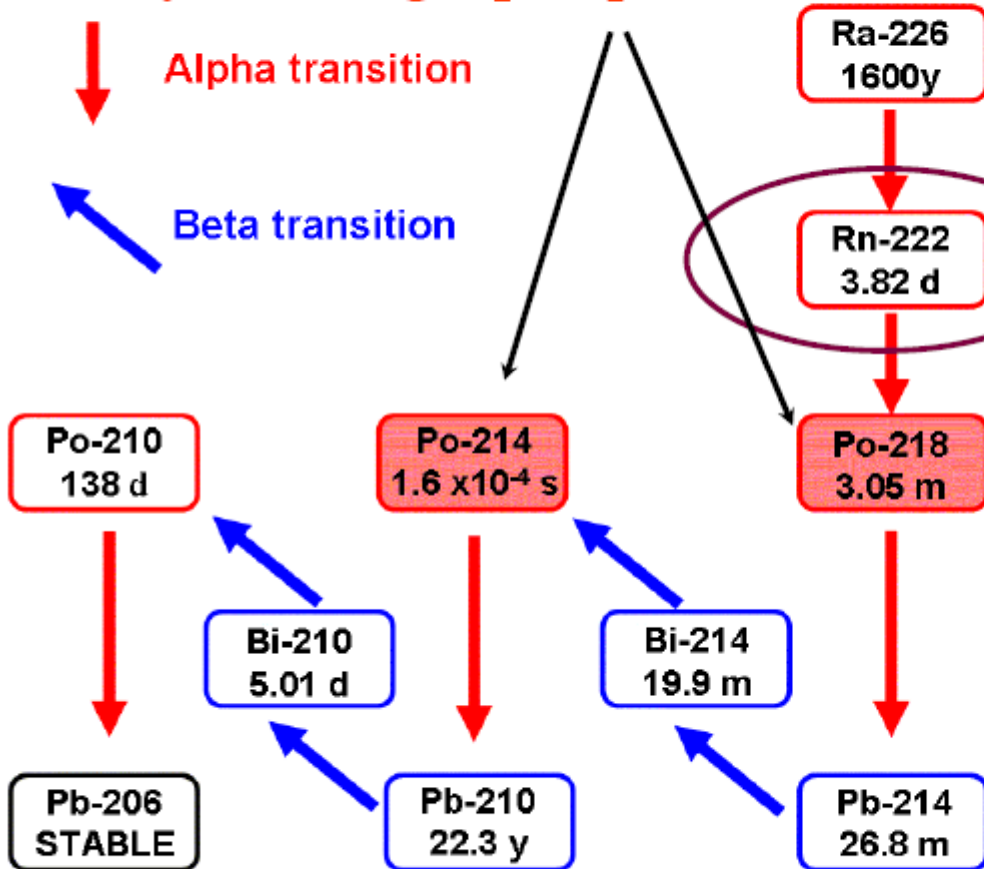
id	im	ih	precip	solar	tpt30	rh10	tpt60	rh60	speed60	dir60grad	iPG	observ
29	8	0	0	0	22.81	57.32	23.55	40.83	1.85	195.29	5	.....
29	8	1	0	0	22.48	60.39	23.32	41.25	1.14	189.03	5	.....
29	8	2	0	0	21.25	56.45	22.44	44.18	1.15	178.36	5	.....
29	8	3	0	0	20.43	63.99	21.98	46.13	0.74	172.86	5	meandering
29	8	4	0	0	19.59	68.09	21.07	49.6	1.64	93.29	5	.....
29	8	5	0	0	19.09	64.84	19.87	56.1	2.96	71.16	5	meandering
29	8	6	0	0.03	18.73	65.7	19.53	57.55	3.24	80.43	6	.....
29	8	7	0	0.07	19.09	64.09	19.34	60.21	2.2	99.98	4	.....
29	8	8	0	0.27	20.91	56.48	20.65	56.65	0.94	94.62	3	.....
29	8	9	0	0.44	23.29	47.81	23	48.55	1.19	83.69	3	.....
29	8	10	0	0.59	25.74	41.92	25.49	43.17	1.83	71.63	3	.....
29	8	11	0	0.68	28.27	30.6	27.89	31.4	2.1	108.27	2	.....
29	8	12	0	0.7	29.93	23.65	29.48	23.54	1.16	174.61	1	.....
29	8	13	0	0.65	30.62	23.73	30.18	23.52	1.23	216.2	1	.....
29	8	14	0	0.52	30.95	25.04	30.45	24.84	0.97	212.04	1	.....

# $^{222}\text{RADON}$

- Naturally occurring radioactive noble gas
- Decay product of  $^{226}\text{Radium}$  in  $^{238}\text{Uranium}$ -series
- Found in all soils, rocks and sediments
- Main source of atmospheric radioactivity (Po,Bi,Pb)
- **Radon is a noble gas** (No deposition, washout or chemical reactions) Only loss mechanism is **radioactive decay**
- The radon half life (3.8 days) is comparable to lifetimes of some important atmospheric species
- Radon is an ideal passive tracer for horizontal transport, vertical mixing processes and soil-atmosphere transport

# How do we measure $^{222}\text{Rn}$ Radon?

By counting alpha particles



Now there are two principal techniques: either measure the air-borne aerosol-bound progeny as complete as possible, mind the **DISEQUILIBRIUM!**

or remove all air-borne progeny from the air sample and wait for new progeny to be produced and measured



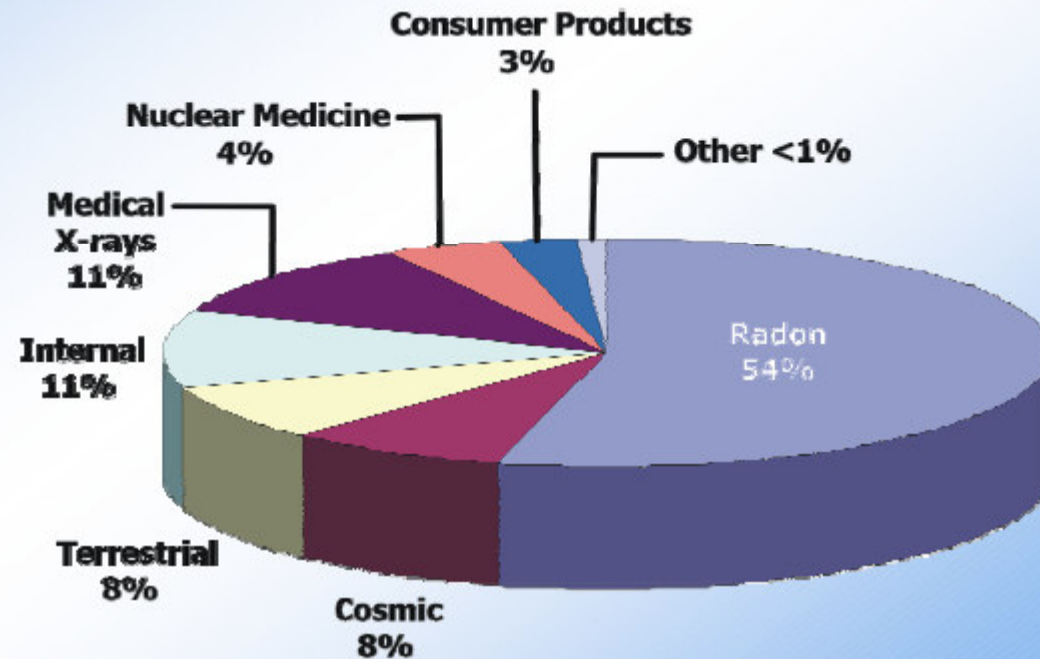
university of  
groningen



Ansto

# RADON contribution to public dose

The **average annual dose** to a person living in the United States is 360 mrem/yr. All natural and man-made sources of radiation contribute to this number.



12/14,15/2005

## IFIN-HH Radon (5 h averaged) at 1 m height - classic method

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

Must be measured at 30 m, above obstacles, with high sensitivity.

**Supplier not yet find for hourly, accurate measurements**

# RADON AND ATMOSPHERIC SCIENCE

- The description of the atmospheric boundary layer on base of measurements of the natural radioactivity is one of the methods used in evaluation of mixing processes in the atmosphere . Observations of atmospheric  $^{222}\text{Rn}$  have been very useful in the evaluation of climate models simulating transport, transformation and removal processes of gases and aerosols . Regional transport models are using also radon for validation .
- The World Meteorological Organization (WMO) established the Global Atmosphere Watch (GAW) Program to investigating the role of atmospheric chemistry in global climate change based on data analysis on many stations. When interpreting results from these stations it is important first to consider how seasonally changing fetch regions and local mixing affect long-term observations at each site, and how comparable observations are between stations in the network in a global context. To these ends hourly radon concentration data are used in conjunction with other observable

## **Diurnal Variations of Radon and Mixing Heights** Along a Coast: A Case Study

S. A. Hsu

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D. J. Bressan

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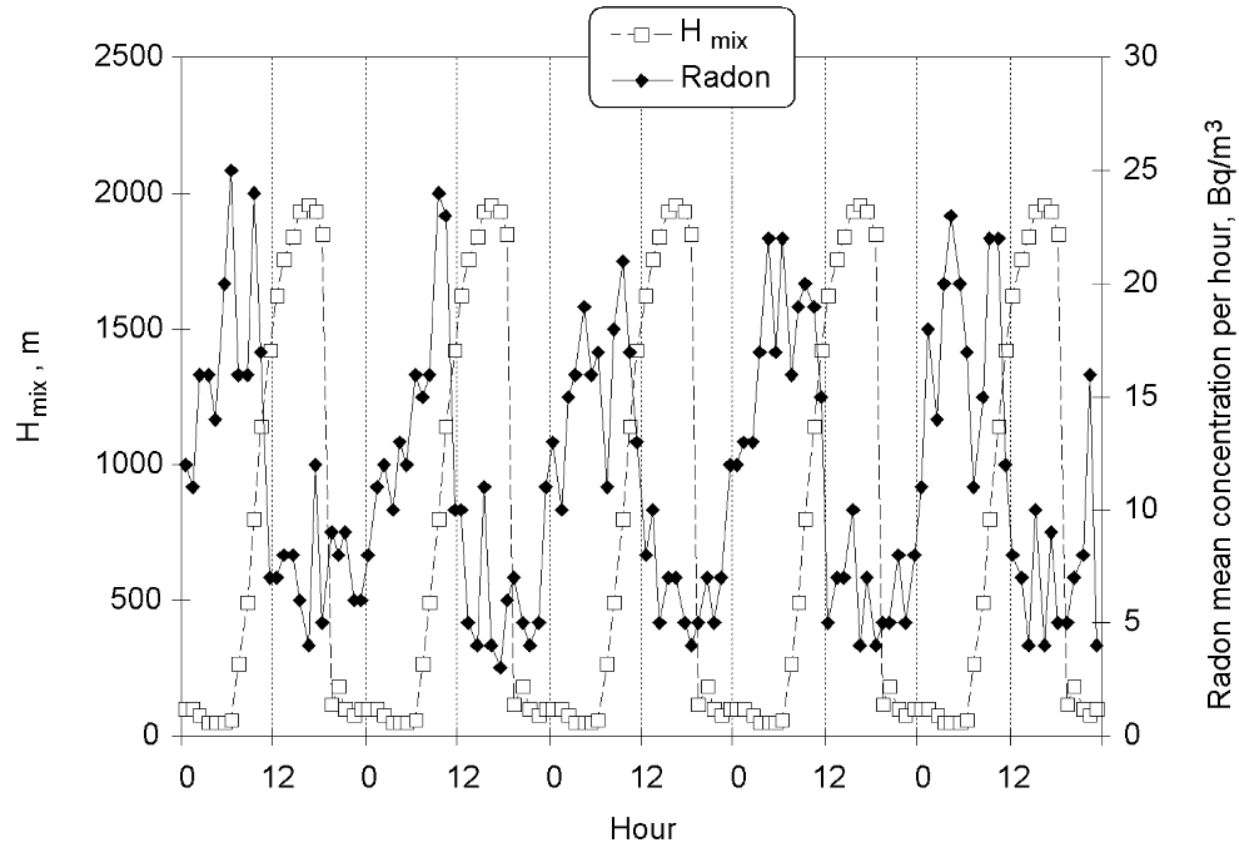
Simultaneous measurements of radon concentration, wind speed and direction near the surface, and atmospheric mixing height within the first kilometer of the planetary boundary layer were made at a beach site on the U.S. Gulf Coast during November-December 1978. Vertical distribution of temperature, humidity, and wind also were available four times daily at approximately 6-hour intervals from two standard rawinsonde stations about 80 km on either side of a beach station along the coast. The mixing height was determined mainly from an acoustic radar (sounder) recorder, but these measurements were supplemented by rawinsonde data. The mixing height as determined by the sounder was verified at least once a day by the rawinsondes before it was correlated with the radon concentration. It was found that after the passage of cold fronts, the meteorology was dominated not only by cold, dry, offshore wind conditions but also by a well-behaved diurnal cyclic variation of both radon concentration

and the mixing height. The radon concentration was found to have an inverse linear dependence on the mixing height, with an unexpectedly high linear correlation coefficient of about  $-0.96$  and conversely to be much less dependent on wind speed than was expected.

**Received 7 December 1979; accepted 10 March 1980;** .

Citation: Hsu, S. A., R. E. Larson, and D. J. Bressan (1980), Diurnal Variations of Radon and Mixing Heights Along a Coast: A Case Study, *J. Geophys. Res.*, 85(C7), 4107–4110.

# Typical result



$H_{mix}$  assessed by CALMET preprocessor using limited direct data



# Diurnal pattern

- A frequently encountered daily pattern is characterized by accumulation of radon in night until the early hours of the morning, followed by a decrease. Nocturnal accumulation of radon occurs under stable atmospheric conditions as the result of the formation of the ground-based temperature inversion; radon mixing takes place in increasingly higher layers in the daily hours
- Nocturnal accumulation of radon indicates the formation of the Nocturnal Stable Layer, where mixing is limited but not completely inhibited.

# emanation

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

# correlations

*Table 2.* Correlation analysis

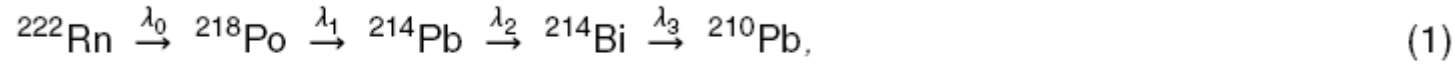
Month and year of sampling	Rn and temperature	Rn and pressure	Rn and humidity
January 03, 04, 05	-0.356	0.213	0.397
February 04, 05	-0.302	0.206	0.297
March 05	-0.258	0.075	0.336
April 05	-0.342	0.293	0.260
May 05	-0.336	0.268	0.429
June 05	-0.291	0.089	0.565
July 02, 05	-0.424	0.163	0.463
August 02, 03, 05	-0.237	0.279	0.284
September 02, 03, 04, 05	-0.185	0.158	0.317
October 02, 04, 05	-0.333	0.248	0.397
November 02, 03, 04, 05	-0.112	0.324	0.387
December 02, 03, 04, 05	-0.190	0.262	0.396

# Data interpretation model (classic)

- In order to obtain quantitative data on the height of the mixing layer ( $h$ ) starting from ground level atmospheric concentrations of radon, a model for the interpretation of these data is necessary. Fontan et al.'s unidimensional box model. The model is based on the mass balance and assumes that:
  - —the flux of radon from the ground is constant;
  - —the radon concentration only varies as a function of vertical stability (i.e. horizontal movement does not alter radon concentration);
  - —mixing of radon in the box is homogenous;
  - —radon in the residual layer is conserved.

## 2 $^{222}\text{Rn}$ decaying chain

We consider the radioactive decay chain of  $^{222}\text{Rn}$  that reads:



where  $\lambda_0$ ,  $\lambda_1$ ,  $\lambda_2$  and  $\lambda_3$  are the decay frequencies equal to  $2.11 \times 10^{-6}$ ,  $3.80 \times 10^{-3}$ ,  $4.31 \times 10^{-4}$ , and  $5.08 \times 10^{-4} \text{ s}^{-1}$ , respectively. Note that we consider a direct transformation of  $^{214}\text{Bi}$  into  $^{210}\text{Pb}$  since the half-life of  $^{214}\text{Po}$  (daughter of  $^{214}\text{Bi}$ ) is very short (164  $\mu\text{s}$ ). Also we consider  $^{210}\text{Pb}$ , that has a half-life of 22.3 years, as an inert scalar with respect to the temporal scales considered here. To increase readability,  $^{222}\text{Rn}$  and its progeny will also be referred to as  $S_i$  where  $i$  is the rank of the daughter in the decay chain from here on, e.g.  $S_0$  and  $S_4$  stand for  $^{222}\text{Rn}$  and  $^{210}\text{Pb}$ , respectively.

In the planetary boundary layer, under horizontally homogeneous conditions with no mean wind and neglecting the transport due to molecular diffusion, the temporal evolution of the mean concentrations  $S_i$  of a radionuclide reads

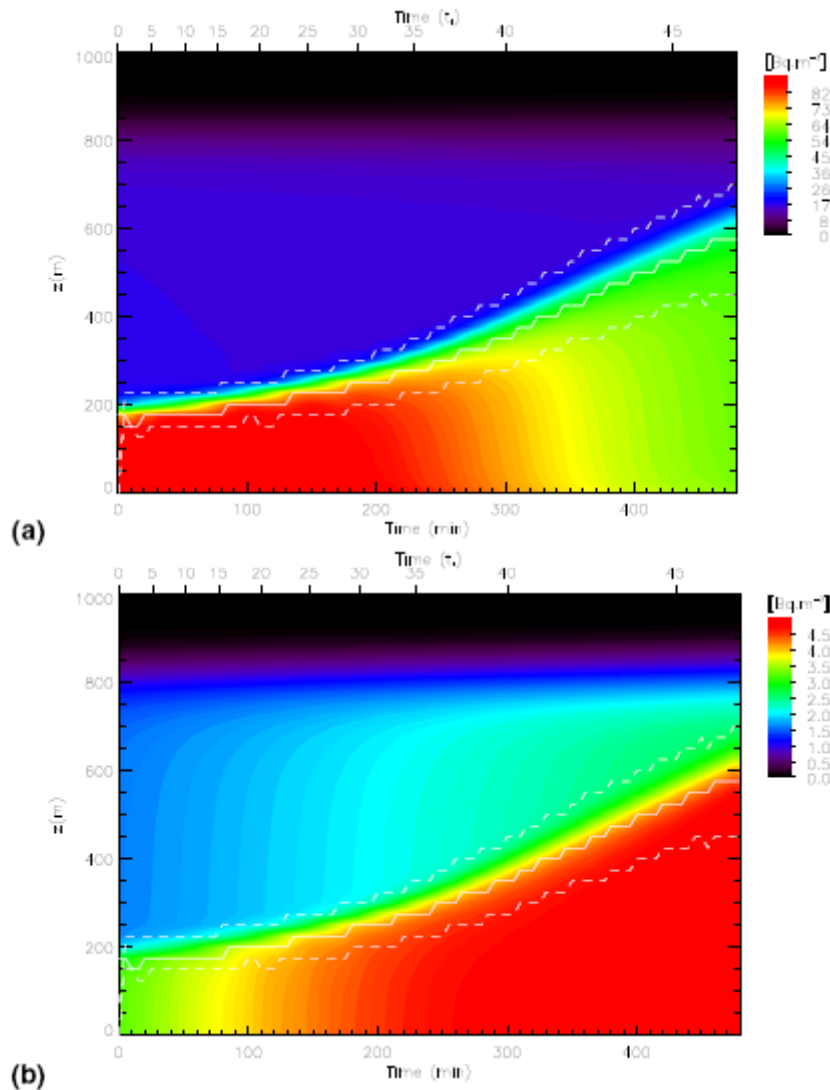
$$\frac{\partial S_i}{\partial t} = -\frac{\partial \overline{w s_i}}{\partial z} + R_{S_i} \quad (2)$$

where the horizontal averages are denoted both by capital letters and overbars whereas the fluctuations of the variables around the horizontal average value are represented by lower case letters. For the chain (1), the radioactive source/sink terms  $R_{S_i}$  are

$$R_{S_0} = -\lambda_0 S_0, \quad (3)$$

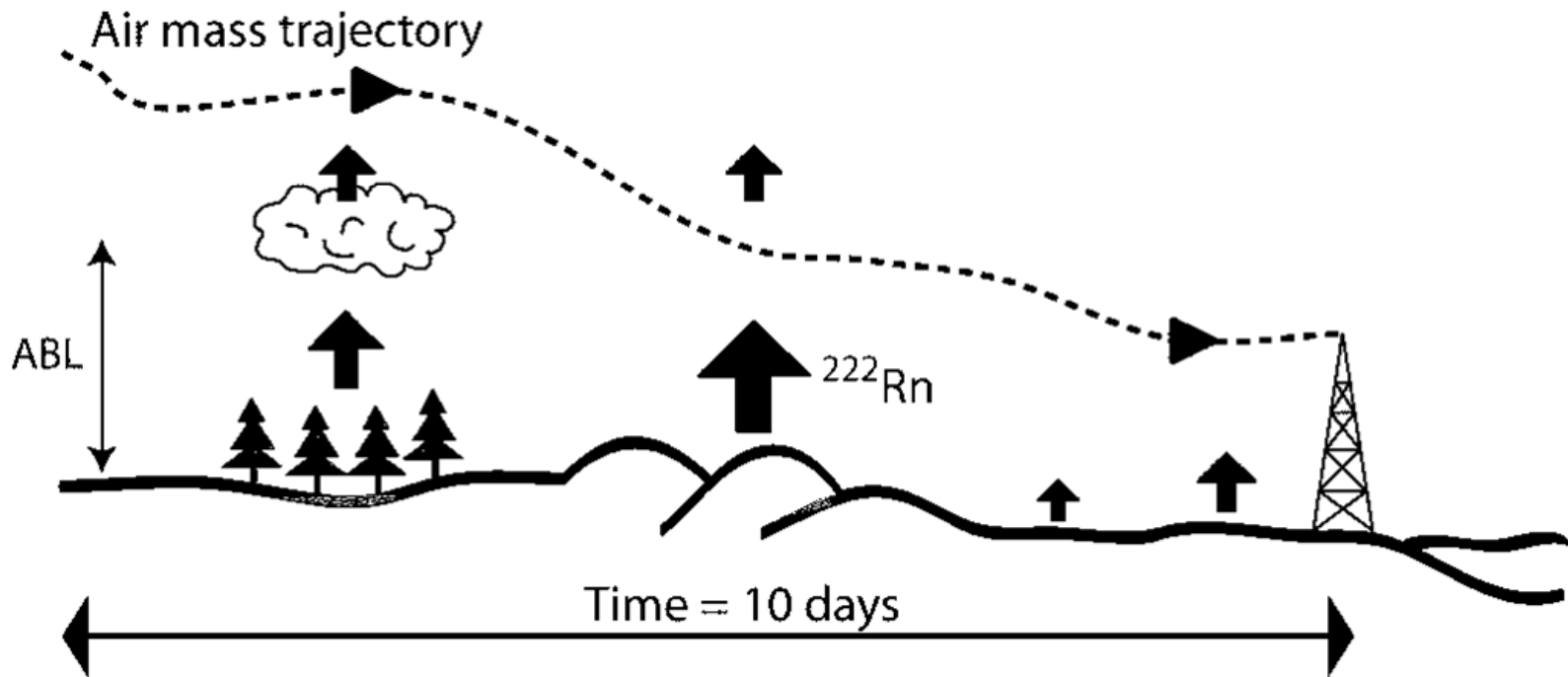
$$R_{S_1} = \lambda_0 S_0 - \lambda_1 S_1, \quad (4)$$

$$R_{S_2} = \lambda_1 S_1 - \lambda_2 S_2, \quad (5)$$



Vertical profiles of **(a)**  $^{222}\text{Rn}$  ( $S_0$ ) and **(b)**  $^{210}\text{Pb}$  ( $S_4$ ) concentrations. The top of the CBL is overplotted with a white solid line and the entrainment layer is located between the dashed white lines. The concentrations are plotted against time in minutes (lower x-axis) and in  $t$  (upper x-axis) where  $t = zi/w$ .

# Foot print



# Clear sky- Radon profile- experimental

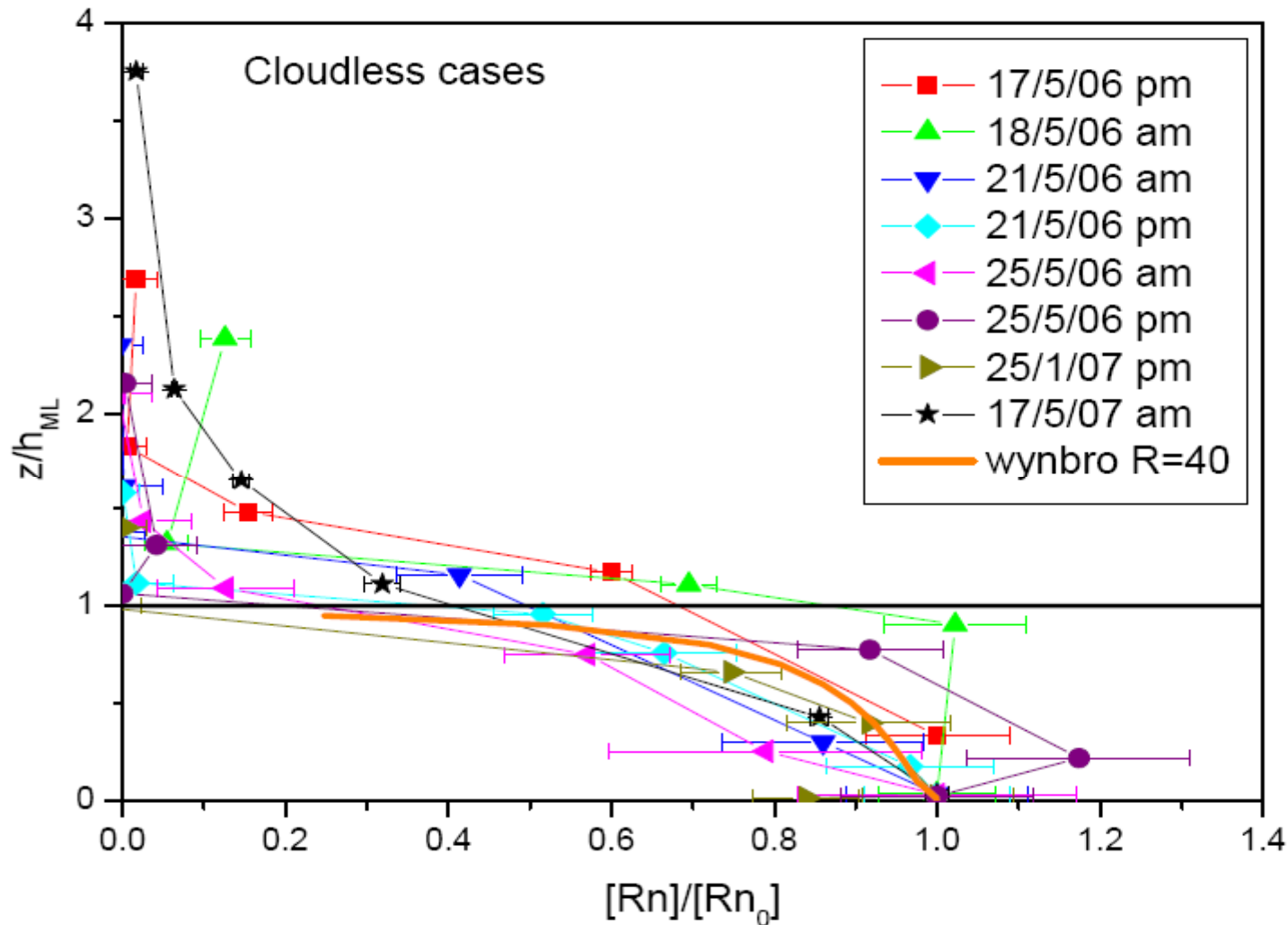
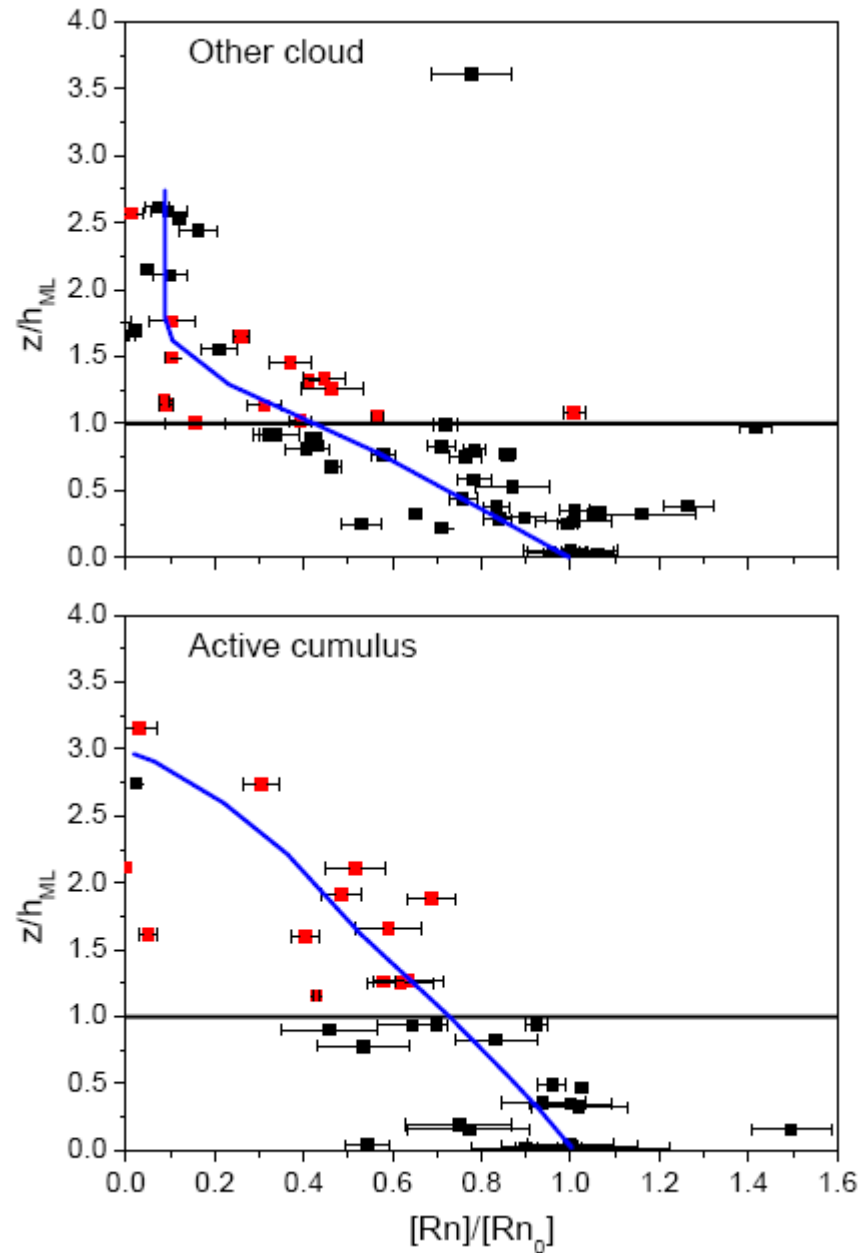


Fig. 4(a)

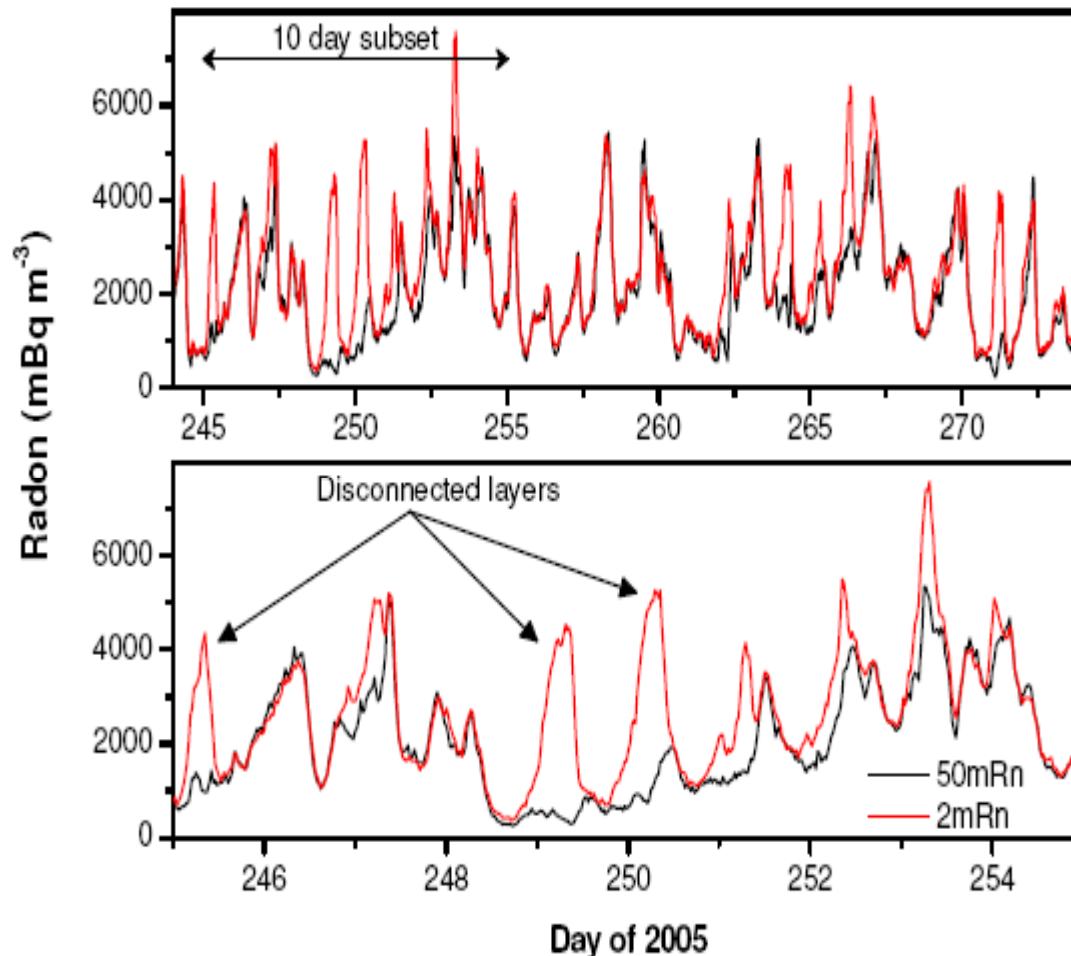
A.G. Williams<sup>1</sup>, W. Zahorowski<sup>1</sup>, S. Chambers<sup>1</sup>, J.M. Hacker<sup>2</sup>, P. Schelander<sup>1</sup>, A. Element<sup>1</sup>, S. Werczynski<sup>1</sup> and A. Griffiths<sup>1</sup> **MIXING AND VENTING IN CLEAR AND CLOUDY BOUNDARY LAYERS USING AIRBORNE RADON MEASUREMENTS 2006**



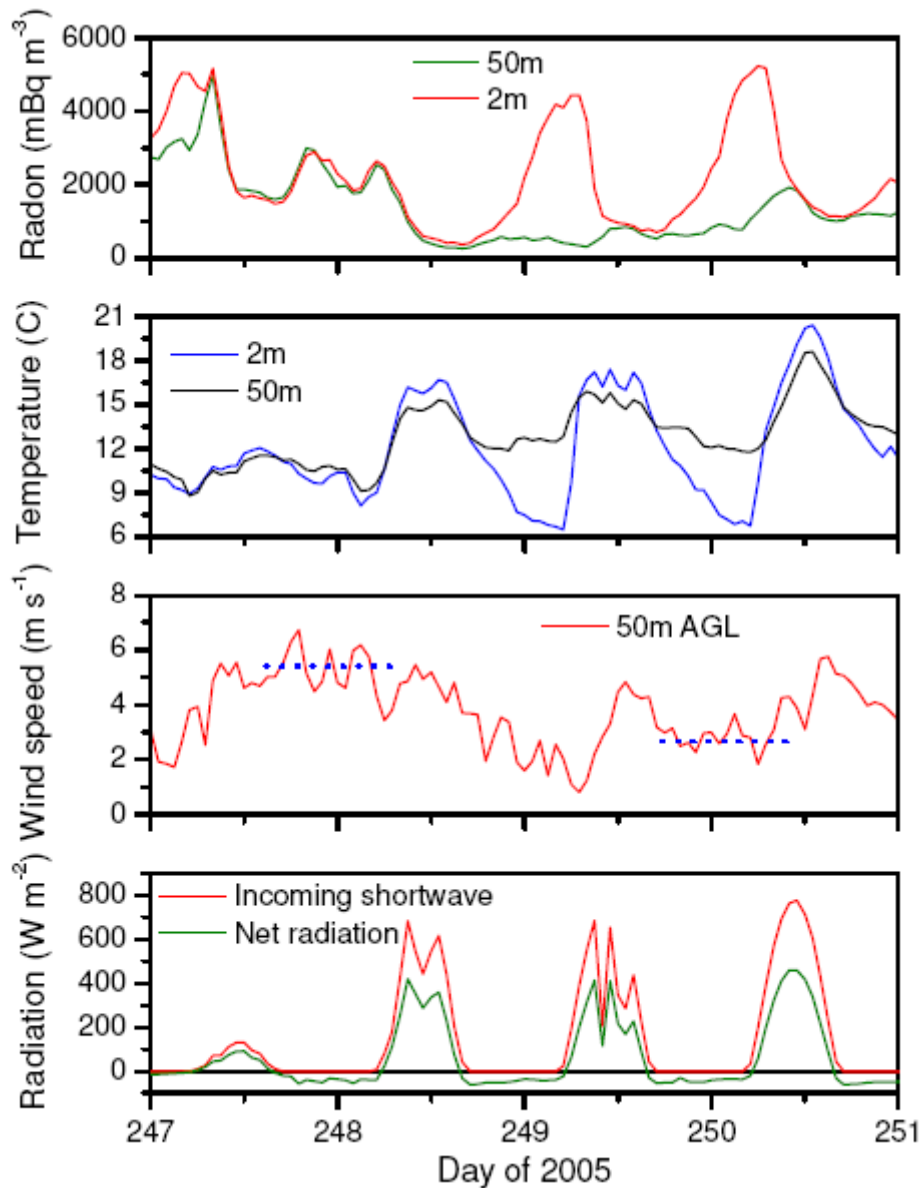
# Radon Profile under clouds



# Space and time -



W. Zahorowski, A.G. Williams, A.T. Vermeulen, S. Chambers, J. Crawford and O. Sisoutham, *Diurnal boundary layer mixing patterns characterised by Radon-222 gradient observations at Cabauw*, [18th Symposium on Boundary Layers and Turbulence](#), Stockholm, Sweden, 9 – 13 June 2008



The above results show that we have a quite complex dependence

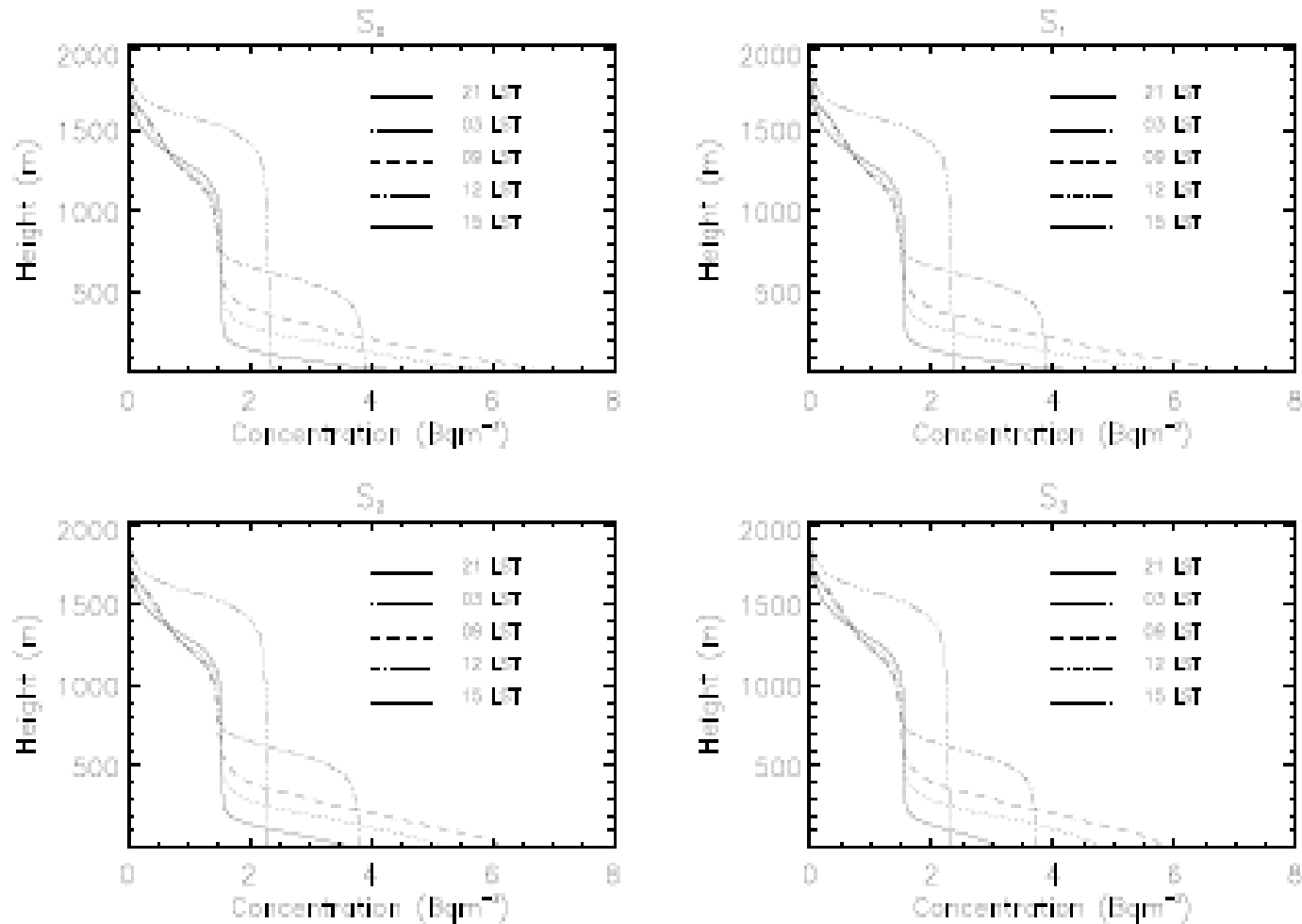
of radon concentration and mixing height and there are many local influences to detect

- source control
- emanation variability
- surface concentration
- Meteorological variables
- mixing height
- simulation of diffusion (advanced models)

Instrument sensitivity

# Simulation of the Wangara experiment

J.-F. Vinuesa, S. Basu, and S. Galmarini *Atmos. Chem. Phys.*, 7, 5003–5019, 2007



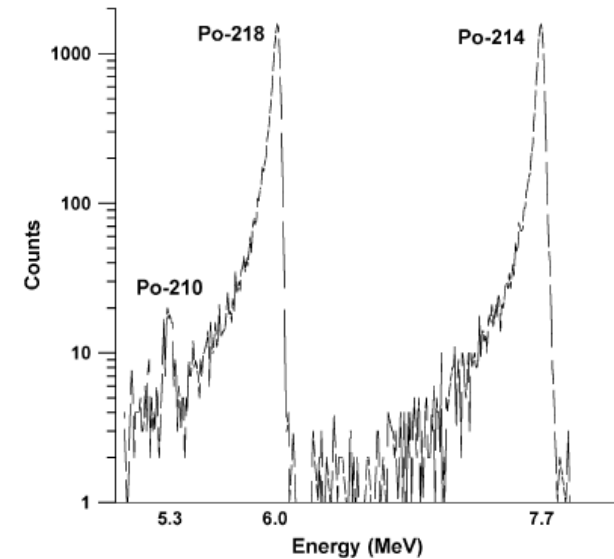
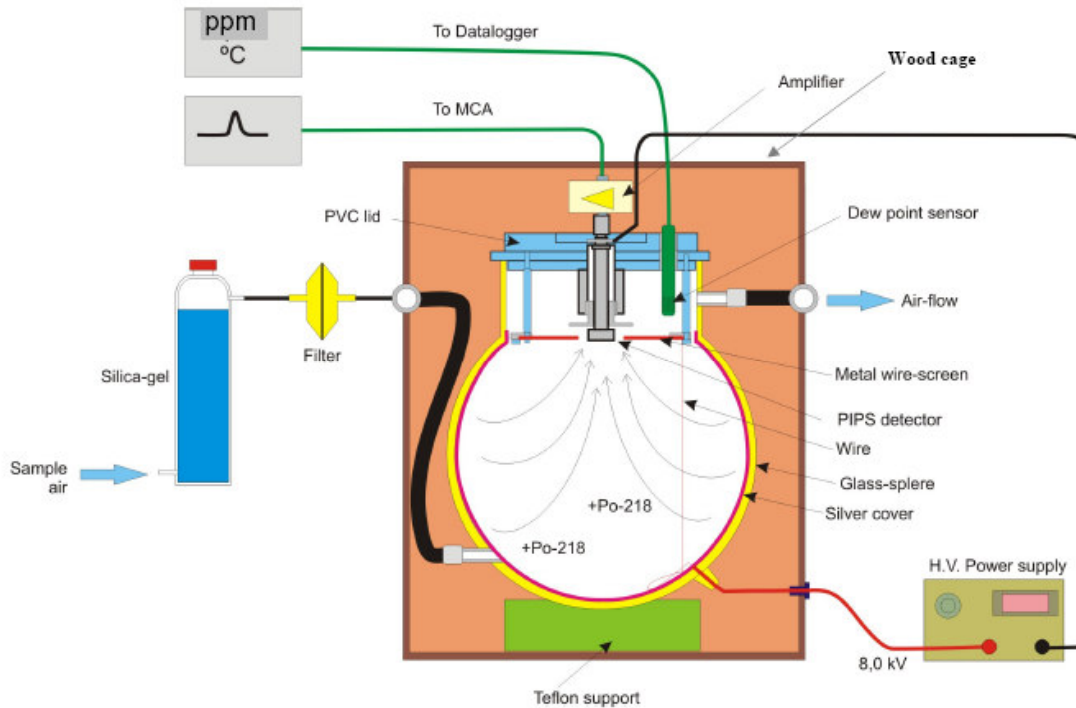
An activity disequilibrium is observed in the nocturnal boundary layer due to the proximity of the radon source and the trapping of fresh  $^{222}\text{Rn}$  close to the surface induced by the weak vertical transport. During the morning transition, the secular equilibrium is fast restored by the vigorous turbulent mixing. The evolution of  $^{222}\text{Rn}$  and its progeny concentrations in the unsteady growing convective boundary layer depends on the strength of entrainment events

# Customer satisfaction

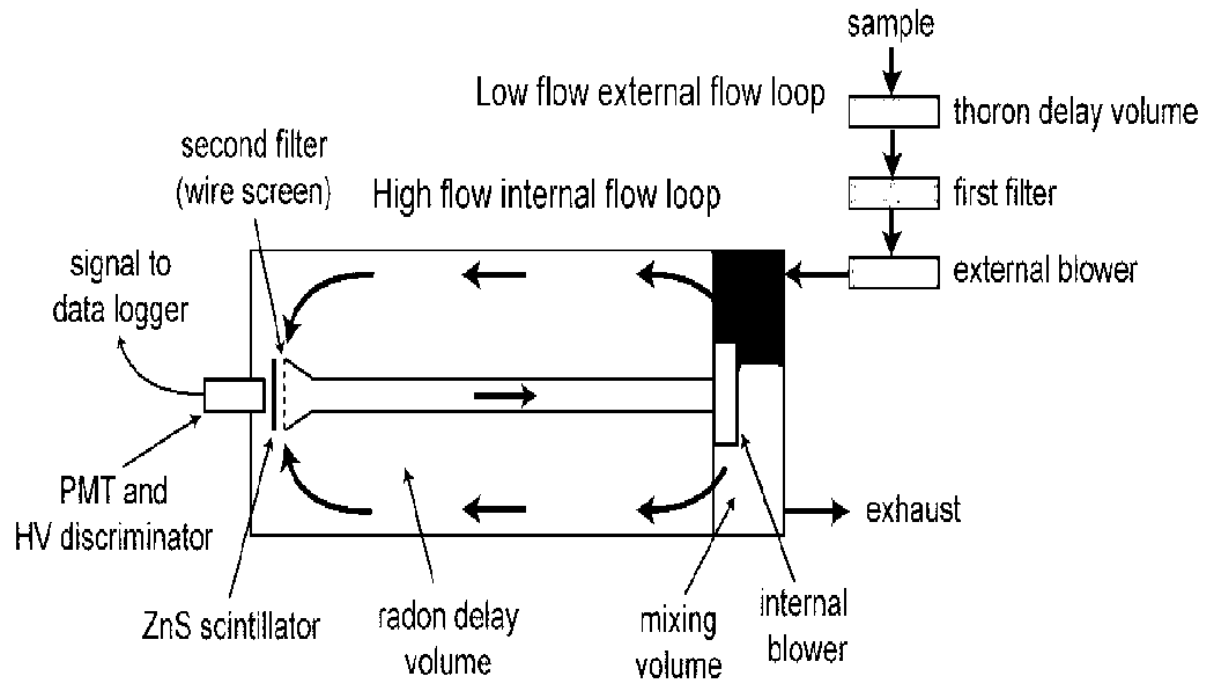
- Producers and distributors want to sell and make profit
- Managers want to report realizations and promote them up
- Scientists want to publish in big journals
- Some of them love to see the results applied in real world
- Budget limitation-global crisis and climate change
- Best instrument- the most difficult problem

A. Vargas Institut de Tecniques Energetiques, Universitat Politecnica de Catalunya, SPAIN

**Scheme of the  $^{222}\text{Rn}$  activity concentration instrument**  
Detection Volume: 20 L (aprox.)



# dual-flow loops two-filter detectors



Zahorowski W.,

# Proposed work plan-synergy

- Eddy covariance- local turbulence, latent & sensible flux, MOL, CO<sub>2</sub> (?) 2010
- Fetch and foot print influences 2011
- Ceilometer- and link with RADO DC- real mixing height 2010
- Best radon instrument under budget 2010
- Data base and correlations >2011
- FLEXPART and MAP3D back trajectories
- Limitations: budget and manpower