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Approximate sequential Bayesian filtering to estimate ^{222}Rn emanation from ^{226}Ra sources from spectra

EMPIR 19ENV01 traceRadon

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***19ENV01 traceRadon** denotes the EMPIR project reference.*

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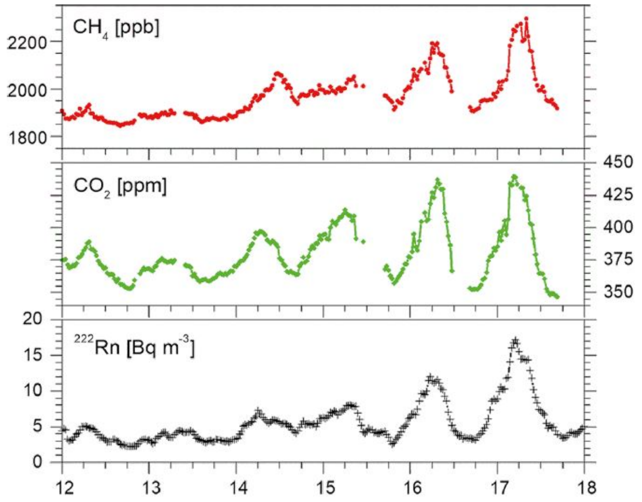
Radon

- ^{222}Rn is an odorless, colorless radioactive noble-gas ($t_{1/2}$ = 3.82 d), which naturally occurs in the environment via the ^{238}U decay chain

- due to diffusion processes, it is accumulated inside of buildings, and is thought to be one of the leading causes for lung cancer [1]
  → EURATOM 2013/58 Basic Safety Standards
  < 300 Bq/m³ inside buildings (1 ^{222}Rn atom per 10^{18} gas molecules)

- Outdoor ^{222}Rn concentration (10 Bq/m³) can be used as a proxy for environmental research ("Radon tracer method" for Green house gases, i.e. [2])
  → **traceradon-empir.eu**

- Reliable (SI-traceable) measurements are necessary



[1]: S. Darby et. al., BMJ, 2005
[2]: C.Grossi et. al., Atmos. Chem. Phys., 18, 5847-5860, 2018

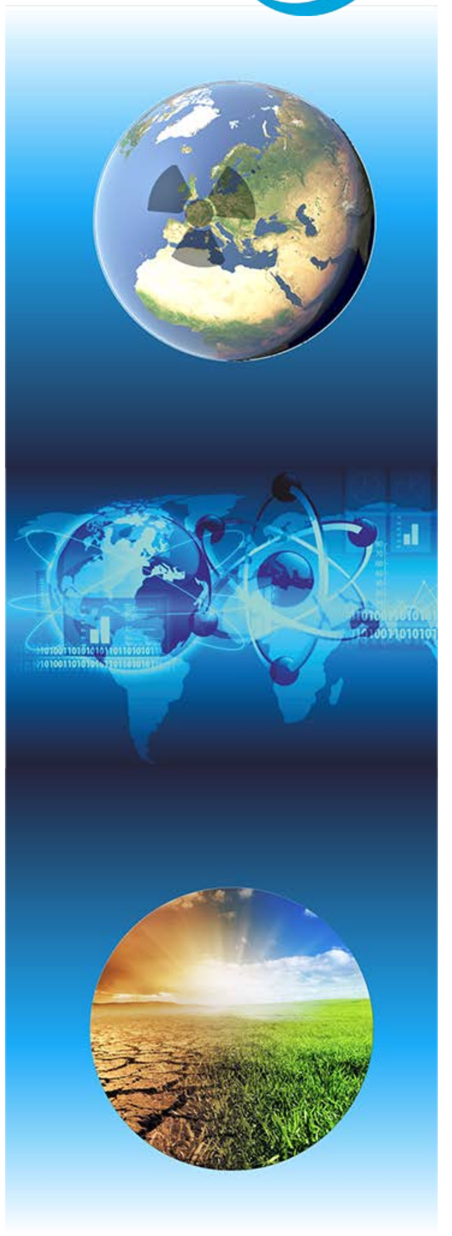
Measurements of ^{222}Rn

- Calibration of ^{222}Rn measurement devices → Reference atmospheres
  - High concentration : gaseous ^{222}Rn standard
  - **Low concentration** : stable (i.e. non-decaying) ^{222}Rn reference atmospheres

How to generate stable atmospheres with known concentration?

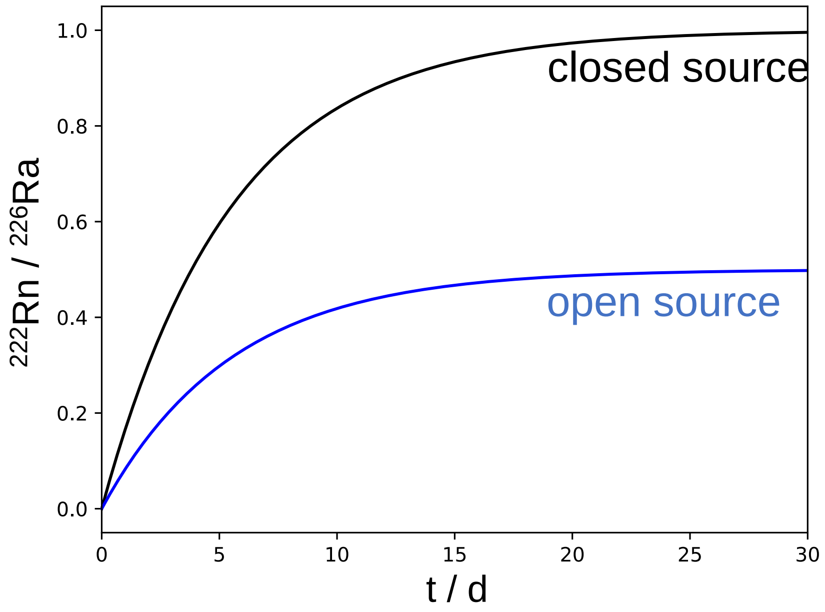
- Use ^{226}Ra in such physical-chemical form that ^{222}Rn is released and is accumulated in some volume (potentially diluted)
- Not all generated ^{222}Rn is released : quantify χ

$$\chi = 1 - \frac{A_{^{222}Rn}}{A_{^{226}Ra}} \qquad [1,2]$$



[1]: D. Linzmaier, A. Röttger, Applied Radiation and Isotopes, 2013
[2]: F. Mertes et. al., Applied Radiation and Isotopes, 2020

^{226}Ra decay chain and kinetics



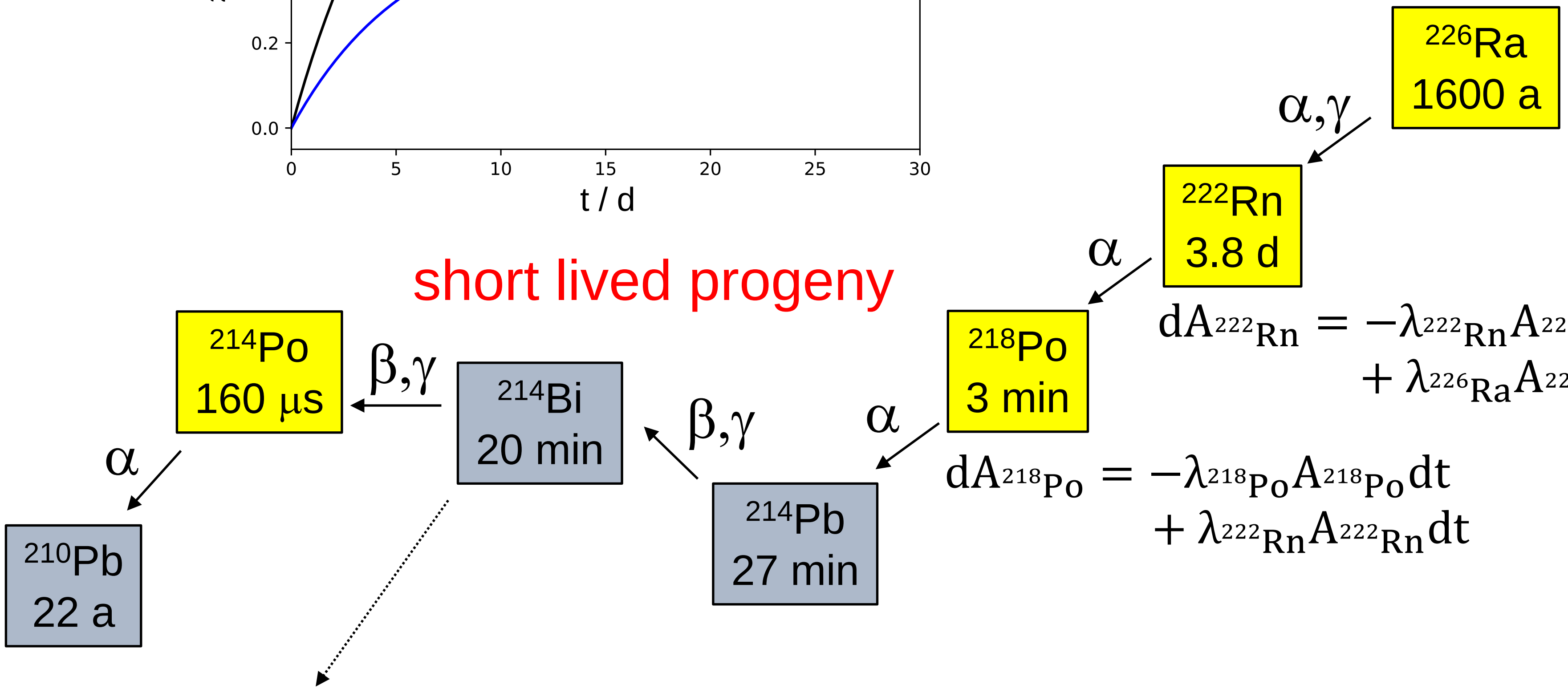
Linear time invariant system

$$dA_{^{226}Ra} = -\lambda_{^{226}Ra} A_{^{226}Ra} dt$$

$$dA_{^{222}Rn} = -\lambda_{^{222}Rn} A_{^{222}Rn} dt + \lambda_{^{226}Ra} A_{^{226}Ra} dt$$

$$dA_{^{218}Po} = -\lambda_{^{218}Po} A_{^{218}Po} dt + \lambda_{^{222}Rn} A_{^{222}Rn} dt$$

short lived progeny



Measuring emanation rate

- χ is a function of environmental parameters (e.g. temperature, pressure, humidity)
  → On-line measurements?

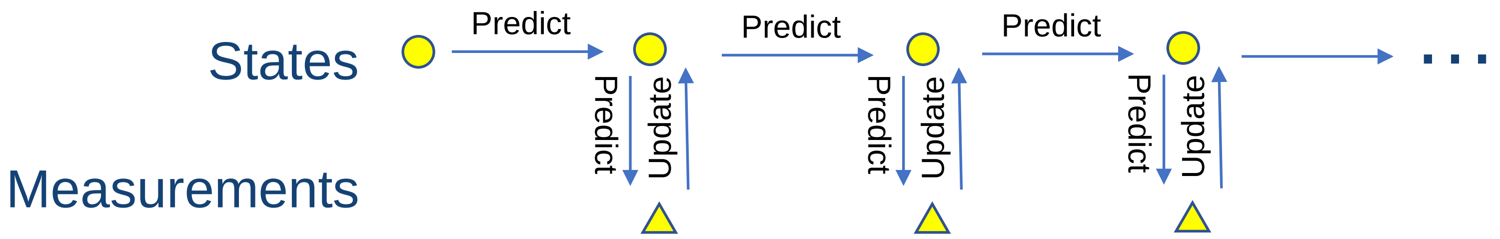
- Previous formula is only applicable under steady-state!

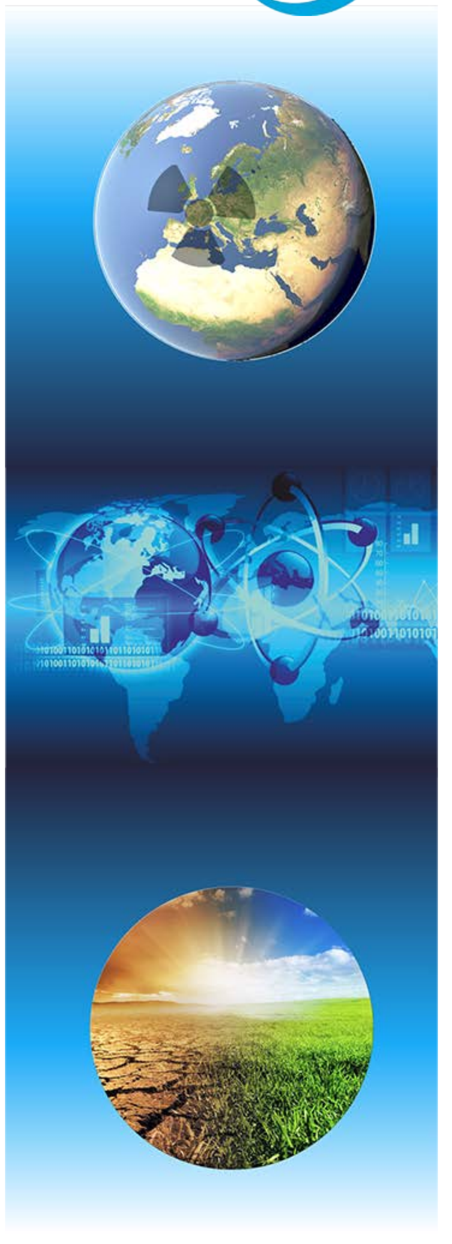
- Sudden change in $\chi(t)$ only appears in the time series of $A_{^{222}\mathrm{Rn}}$ as its convolution with the solution of the linear time-invariant system of ingrowth differential equations

- Estimating the emanation rate $\eta(t)$ (released ^{222}Rn atoms per unit time) from a time-series of measurements is an inverse-problem

→ Bayesian sequential filtering on state-space x(t)

$$\mathrm{x} = \left[A_{^{222}\mathrm{Rn}} \quad A_{^{226}\mathrm{Ra}} \quad \eta \quad \frac{d\eta}{dt} \right]^{\mathrm{T}}$$





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State space model

- General approach: Model $\eta(t)$ as a stochastic process, which drives the system of DEs

$$dA_{^{222}Rn} = -\lambda_{^{222}Rn} A_{^{222}Rn} dt + \lambda_{^{226}Ra} A_{^{226}Ra} dt - \lambda_{^{222}Rn} \eta(t) dt$$

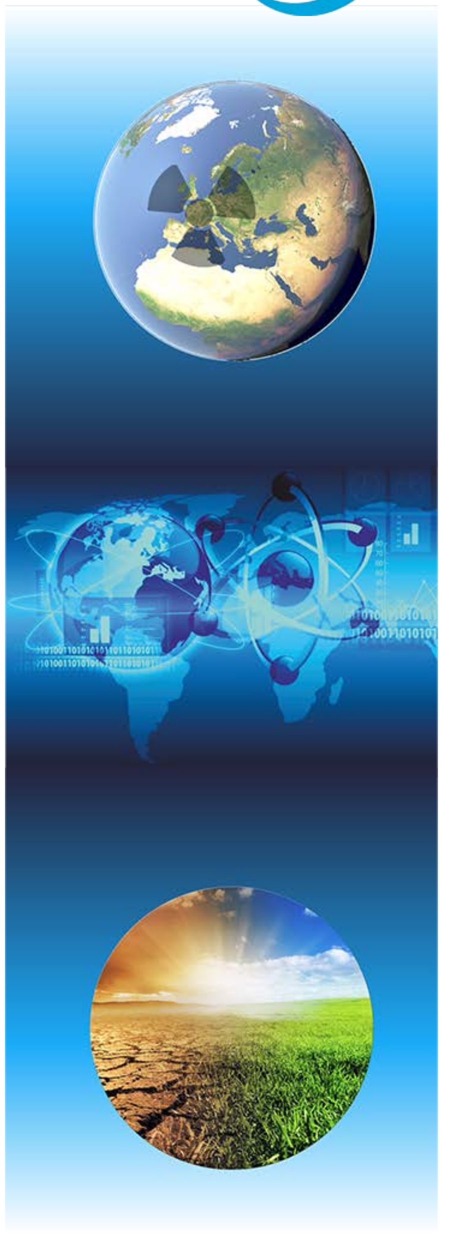
change = - decay + ingrowth - outflow

- Since $\eta(t)$ is completely latent, we need to account for the "unknown-ness" of it, through allowing it to have some randomness → Use a stochastic process, but matter of prior assumptions (e.g. comparable to placing a prior on "allowed" function types that $\eta(t)$ must match)

- For example, restrict to somewhat smooth behaviour (e.g. continuous 1st derivative)

$$\frac{d^2\eta}{dt^2} = -\gamma \frac{d\eta}{dt} + d\beta$$

Brownian motion / Random increments
Power spectral density Q (unknown)



Inference steps - Prediction

1. Predict new state value at time instant t from prior measurement $x = \left[A_{^{222}Rn} \quad A_{^{226}Ra} \quad \eta \quad \frac{d\eta}{dt} \right]^T$

$$dx = Fxdt + Ld\beta \qquad x(t) = e^{F(t_0 - t)} x(t_0) + \int_{t_0}^{t} e^{F(t-\tau)} L d\beta_\tau$$

Itô stochastic integral

$$p(x_t | x_0) \propto N\left(\Psi(t, t_0)\mu_{x_0}, \Psi(t, t_0)\Sigma_{x_0}\Psi^T(t, t_0) + W\right)$$ [1]

$$\Psi(t, t_0) = e^{F(t - t_0)} \qquad W = \int_{t_0}^{t} e^{F(t-\tau)} L Q L^T e^{F^T(t-\tau)} d\tau$$

We have

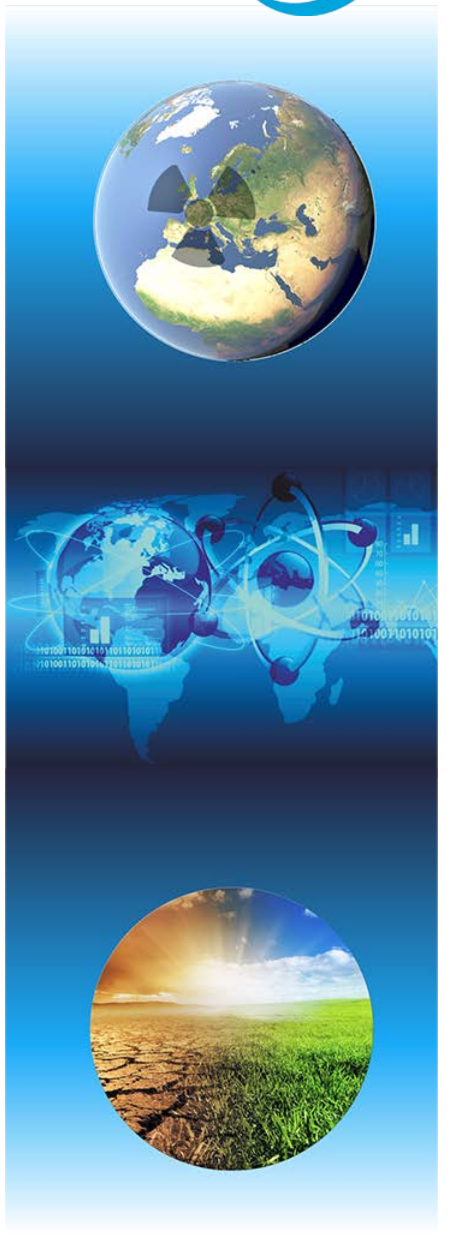
$$F = \begin{bmatrix} -\lambda_{^{222}Rn} & \lambda_{^{222}Rn} & -\lambda_{^{222}Rn} & 0 \\ 0 & -\lambda_{^{226}Ra} & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 0 & -\gamma \end{bmatrix}$$

for with the above can explicitly be calculated (analytically), i.e. using the Jordan form.

x(t) is a Gaussian process.

Radioactive system

Assumptions about η



[1]: Simo Särkkä, Arno Solin, Applied Stochastic Differential Equations, 2019, Cambridge University Press

Inference steps – Correction

2. Correct predicted state based on a measurement

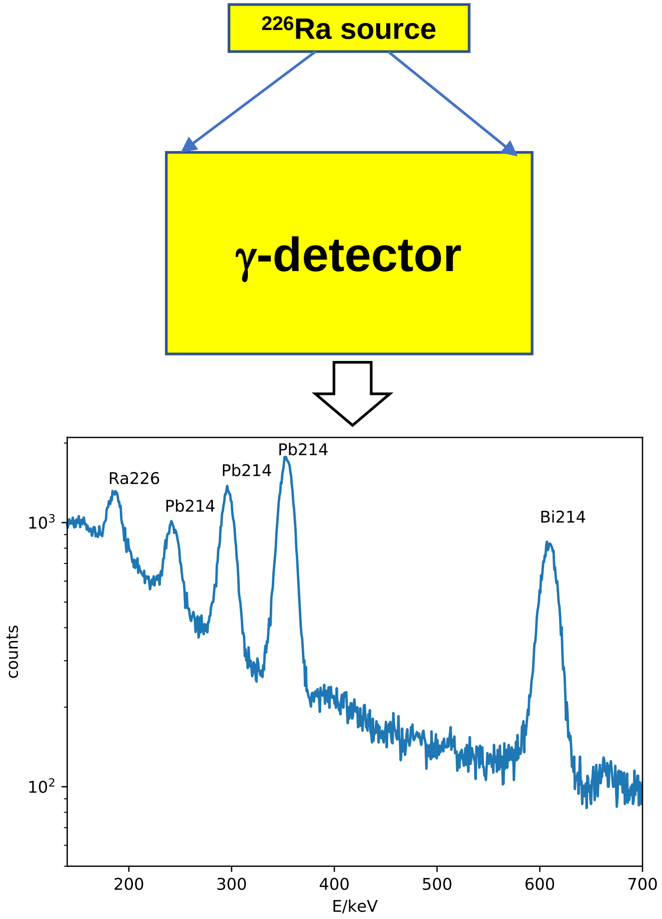
$$x = \left[A_{^{222}Rn} \quad A_{^{226}Ra} \quad \eta \quad \frac{d\eta}{dt} \right]^T$$

- Measure $A_{^{222}Rn}$ (and possibly $A_{^{226}Ra}$) through measuring a spectrum (i.e. γ-ray spectrum)

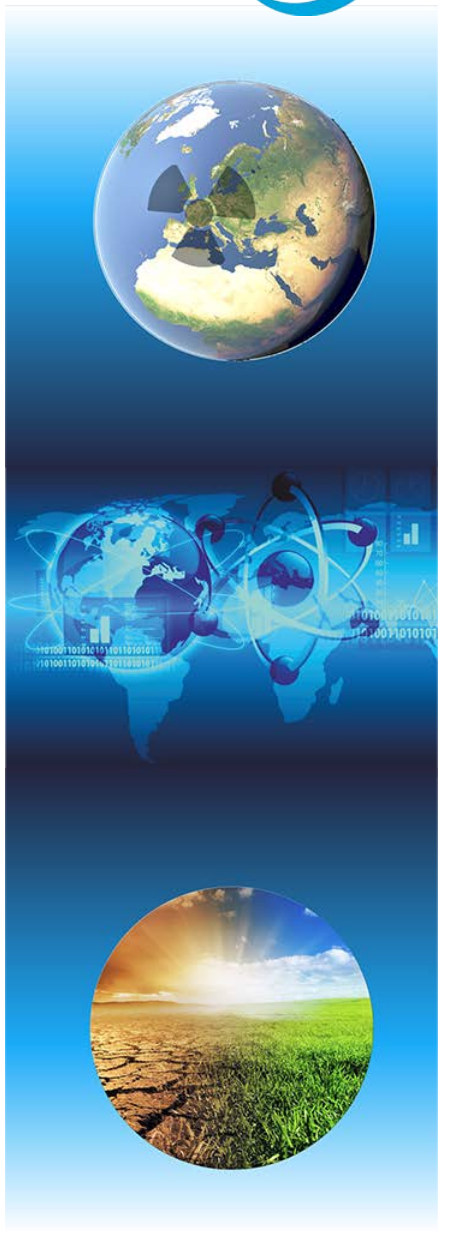
- However, spectral data acquisition takes time (typical integration time 10^4 s)
  e.g. integrates the state, over a stochastic process

  thus

$$y(t, l_t) = H \int_0^{l_t} x(t+\tau)d\tau$$



Can be tricky to solve, since it results in a Riemann integral over a stochastic Itô integral



Inference steps – Correction

- It can be shown that, in these models, the measurements are also a Gaussian process, where

$$p(x_t, y_t) \propto N\left(\begin{bmatrix} \mu_{x_t} \\ K_{l_t}\mu_{x_t} \end{bmatrix}, \begin{bmatrix} \Sigma_{x_t} & \Sigma_{x_t} K_{l_t}^T \\ K_{l_t}\Sigma_{x_t} & K_{l_t}\Sigma_{x_t}K_{l_t}^T + J_{l_t} + R_t \end{bmatrix}\right)$$

$$K_{l_t} = H \int_0^{l_t} e^{F\tau} d\tau$$

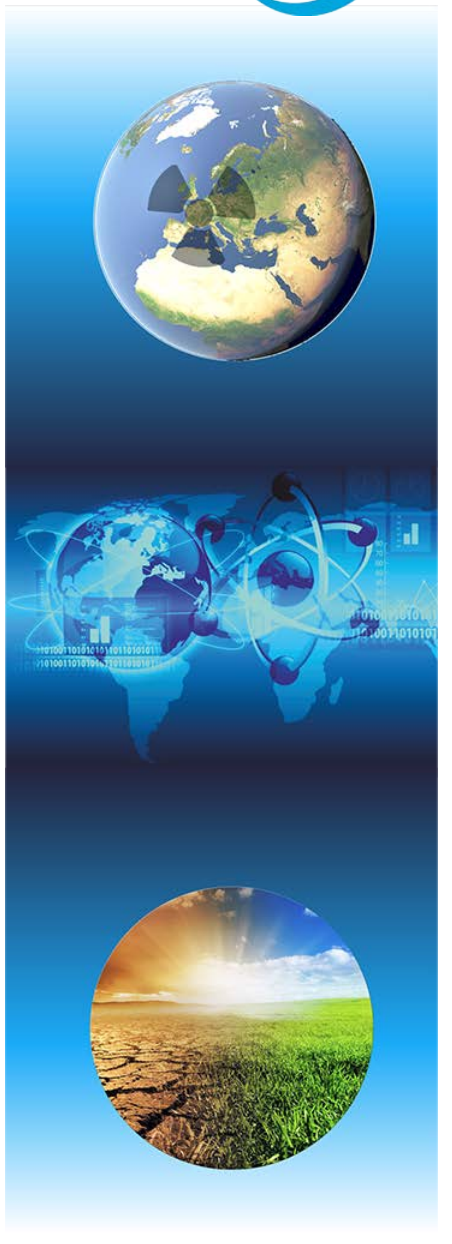
$$J_{l_t} = H \int_{-l_t}^{0} \int_0^{\tau} \int_0^{\tau} e^{Fa} L Q L^T e^{F^T b} \, da \, db \, d\tau \, H^T$$

$$R_t$$

Propagation factor to account for integrating

Additional variance from integrating the stochastic part of the process (scary, in our case symbolically. Numerical Algorithms are available)

Measurement noise Variance. Estimated from observed counting statistics (e.g. $\sigma = \sqrt{N}$)



Improvement

- Q (power spectral density of driving Brownian motion process) is a parameter of the model. Tuned for performance of filter (based on likelihood of collected data):

  high Q : faster reaction to steep changes, high noise in filter output

  low Q : inability to react to steep changes, low noise in filter output

- not ideal, we need the best of both worlds : Switching linear dynamical system! (SLDS)

  Bonus: probabilistic change-point detection

Idea is to use multiple indexed filters and calculate the probability for the active one (based on likelihood of its prediction)

but for SLDS, the posterior is a mixture of Gaussians, which can not be analytically computed. Exponential explosion of mixture components.

Algorithmic approximation : Collapse mixture to a smaller but ideally fitting mixture [1] (i.e. through approximation of their Kulback-Leibler divergence [2])

[1] D. Barber, Journal of Machine Learning Research, 2006
[2] A. R. Runalls, IEEE Transactions on Aerospace and Electronic Systems, 43, 2007

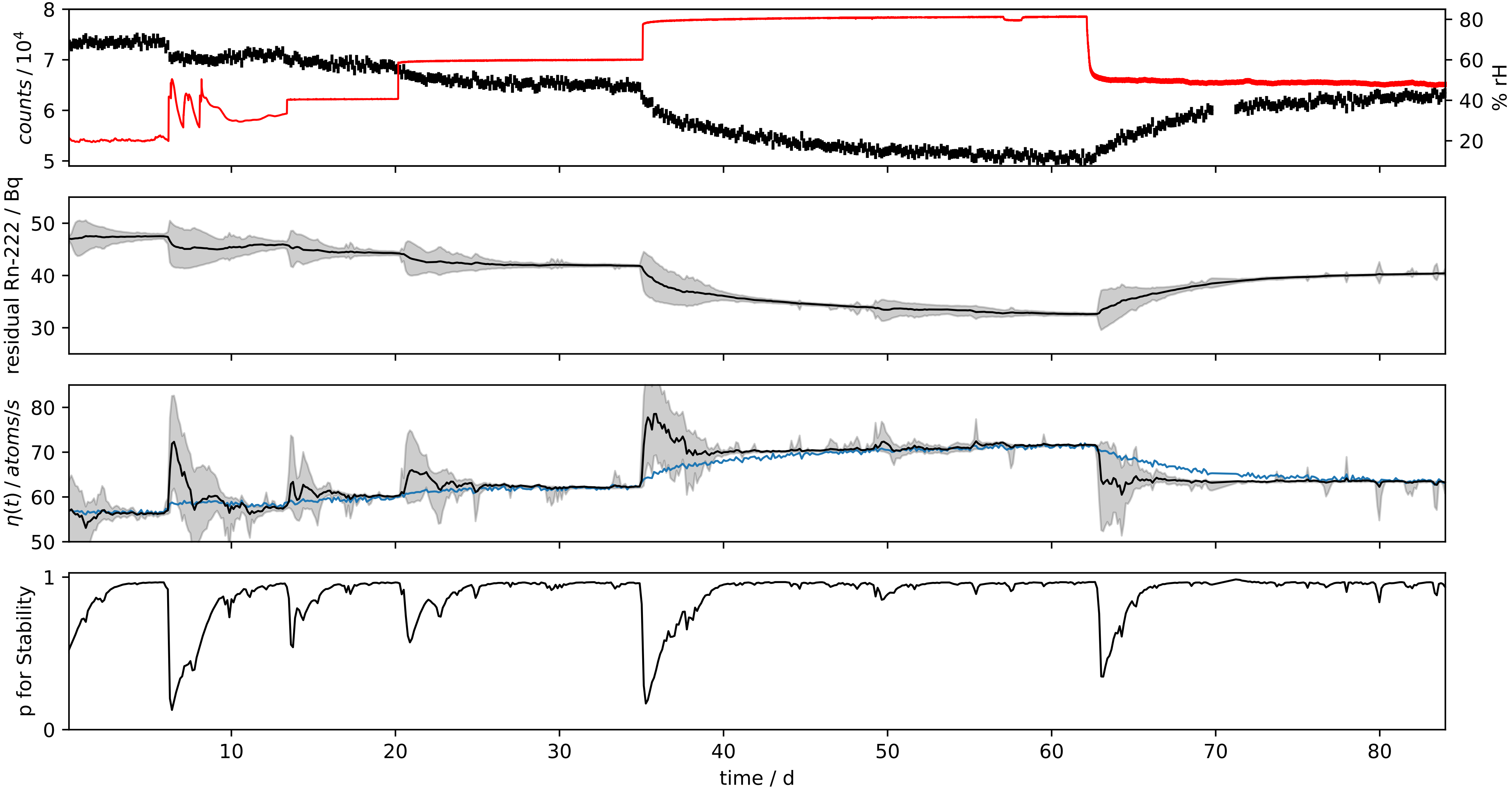
Example for a 2-Filter setup

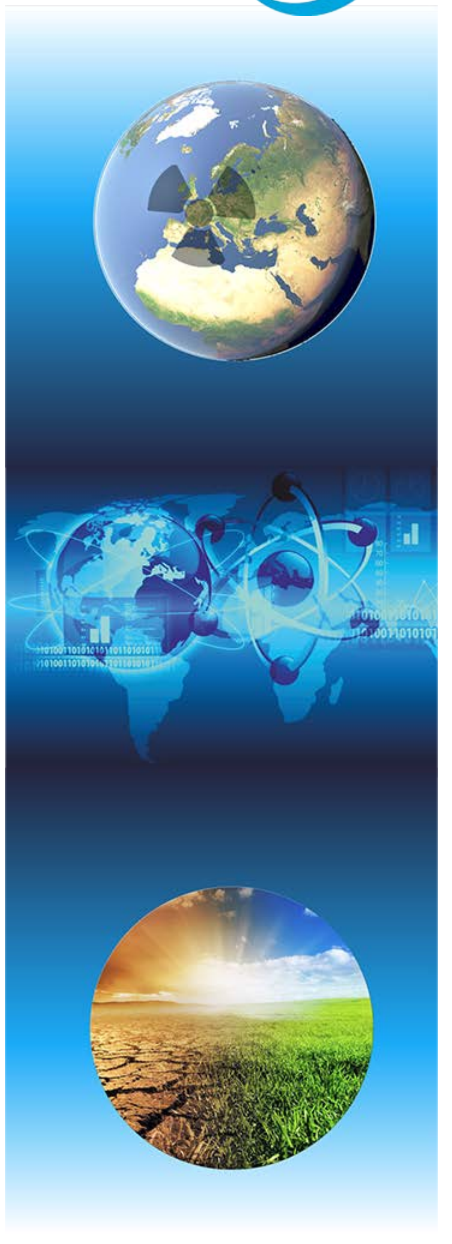
- Expect $\eta(t)$ to be constant over prolonged times but then suddenly change

- Model proposal for a specifically recorded time-series:
  - Filter 1: “0 Variance Brownian Motion” → completely deterministic dynamics
  - Filter 2: High Variance of Brownian Motion → can adapt quickly to change in $\eta(t)$
  - Posterior is mixture of both filters, weighted by the likelihood of their predictions
    → This is also the probability for stable regimes in the time-series
  - Inference in such systems is well reported on in literature, e.g. [1]
  - Tuning of model parameters through maximum likelihood (e.g. which filters explain the time-series best, only approximately possible)
  - Implemented in C++ for speed (using EIGEN Library for Cholesky decomposition)

[1] D. Barber, Journal of Machine Learning Research, 2006

Example Filter output





Conclusion

- A physically motivated model to infer the ^{222}Rn emanation behaviour based on continuous measurements of residual ^{222}Rn was developed

- Analysis of a time-series over 85 days shows reasonable estimation of the emanation and its uncertainty

- To account for the integrating measurement behaviour, the algorithms reported on in literature were extended to incorporate this, by deriving the statistical properties of this system from theory

- A first step towards establishing sequential Bayesian inference in radioactivity analysis, which are currently not widespread (even though it is a prime example of a LTI system)

- Possibility for on-line operation is given

- For now, only filtering is done (no backwards pass through the data, e.g. every inferred point only depends on data that was available up to that time)