Assessment of regional atmospheric transport model performance using ^{222}Radon observations

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Assesement of regional atmospheric transport models

- Statistical inversions provide a way of estimating greenhouse gas (GHG) fluxes and emissions from measurements of atmospheric GHG concentrations, independent from national reportings or inventories.
- Transport models are a central part of theses inversions and quantitative knowledge of their uncertainties is a prerequisite for the inversion performance as any unaccounted uncertainty or systematic error in the inversion system directly translates to errors in the flux estimates.
- The Stochastic Time Inverted Lagrangian Transport STILT is one example of a regional transport model. STILT is routinely used in inversions. It is also available as an online tool to support analysis of atmospheric CO_2 at ICOS stations.
- In this study, we explore possibilities to assess the ability of the model to correctly simulate the diurnal variation of boundary layer transport by comparing model results with observations of atmospheric Radon activity concentration.

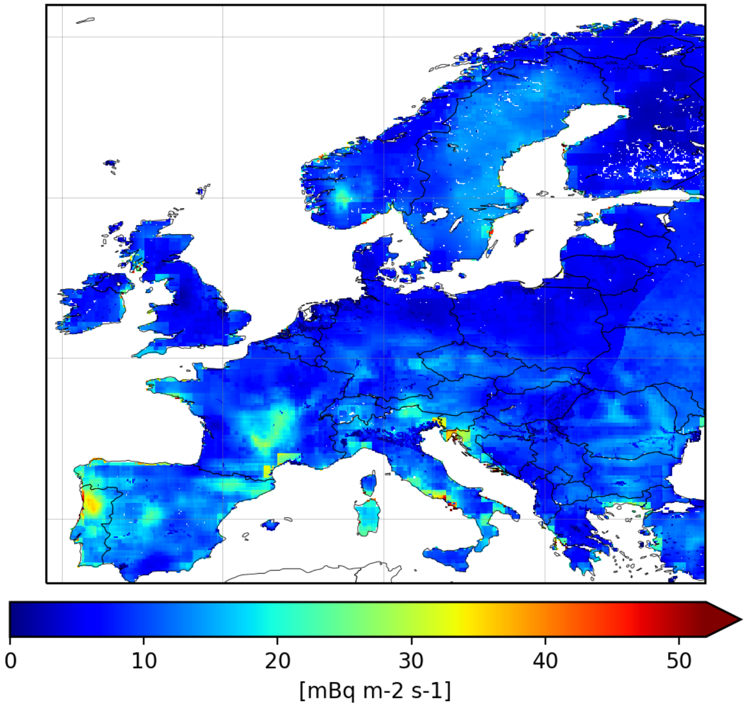
Radon as tracer for atmospheric transport

- Radon has a fairly homogeneous surface source (compared to greenhouse gas fluxes)
  - Radon is a decay product of Uranium, a component of all natural soils
  - Exhalation rate of Radon depends on soil type and permeability
  - Oceanic Radon fluxes are orders of magnitude smaller than fluxes from soils
- Radon has a well-defined sink
  - Radioactive decay with a half-life of 3.82 days
    - longer than turbulent time scales (~conservative tracer in atmospheric boundary layer)
    - short enough to build up a vertical gradient in the troposphere
- Due to these characteristics, Radon can be applied in atmospheric mixing studies and also as tracer for the evaluation of transport model performance.

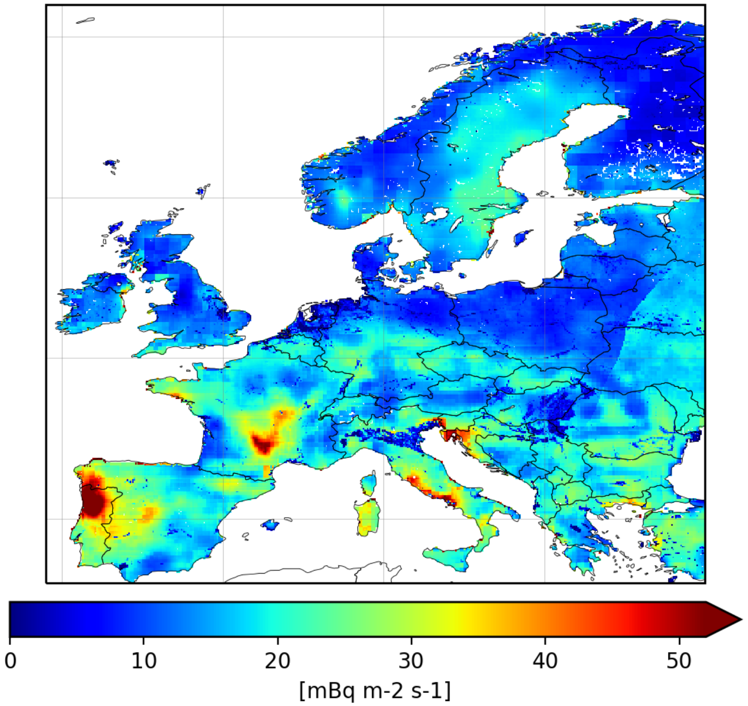
However, this requires knowledge of the continental Radon exhalation rate.

Radon flux map for Europe

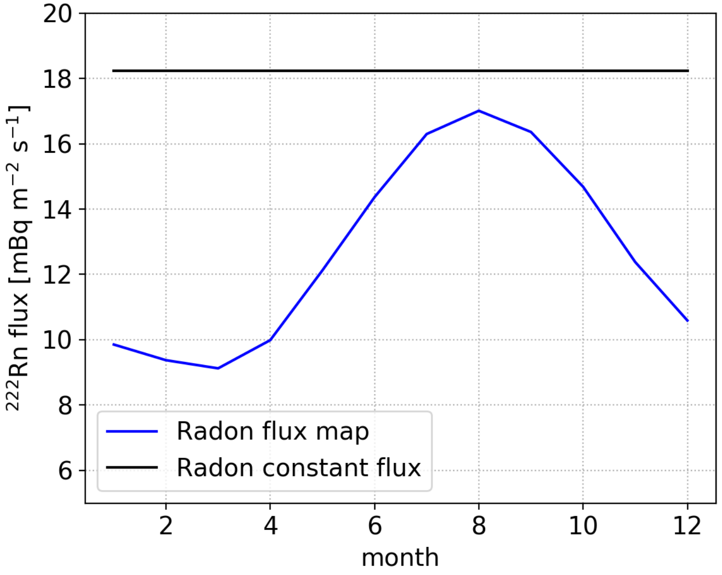
^{222}Rn exhalation rate Januar



^{222}Rn exhalation rate July



Average ^{222}Rn exhalation from European land surface



The black line indicates a constant Radon flux (1 atom m^{-2} s^{-1}) that is often used in global transport modelling studies.

- Climatological Radon flux map based on Karstens et al. (2015)
  - Radon source ~ Uranium content → spatial variations
  - Soil texture, porosity → spatial variations
  - Soil moisture (land surface reanalysis products) → temporal variations

Radon measurements in the ICOS atmospheric station network

- Recommended measurement component

- Four tower stations selected here for model comparison:

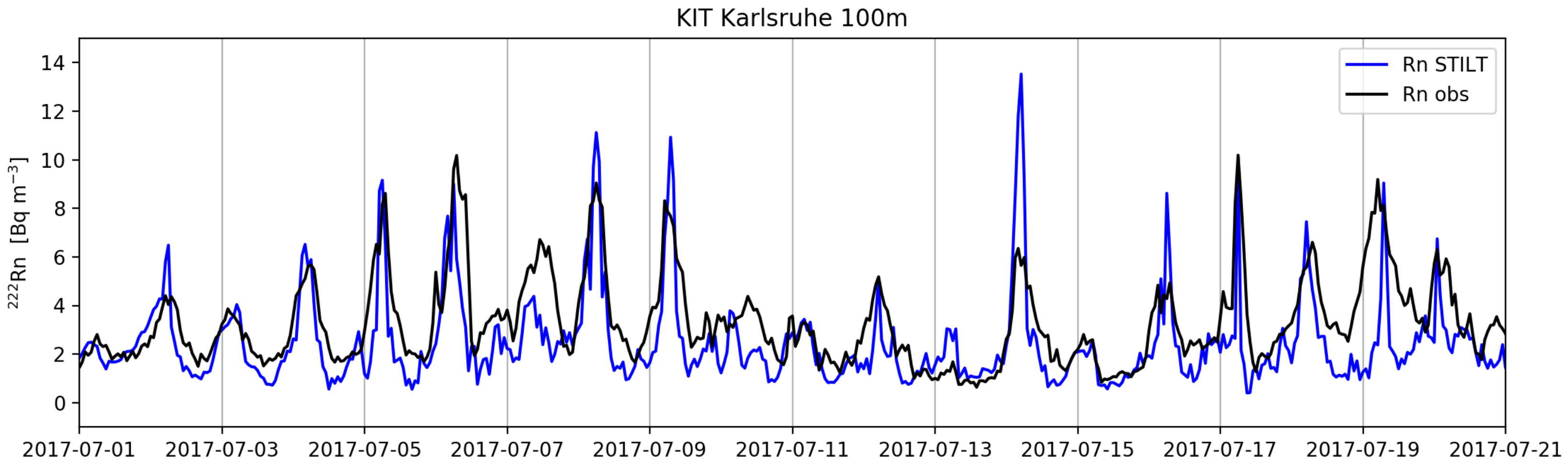
- Gartow (132 m a.l.g., ^{214}Po: HRM*)
- Lindenberg (99 m a.g.l., ^{214}Po: HRM)
- Trainou (180 m a.g.l, ^{222}Rn: ANSTO)
- KIT Karlsruhe (200 m, 100 m, ^{214}Po: HRM and 30 m ^{214}Po: HRM and ^{222}Rn: ANSTO)

- All HRM ^{214}Po data corrected for line loss (Levin et al., 2017) and disequilibrium and brought to ANSTO scale (Schmithüsen et al., 2017)

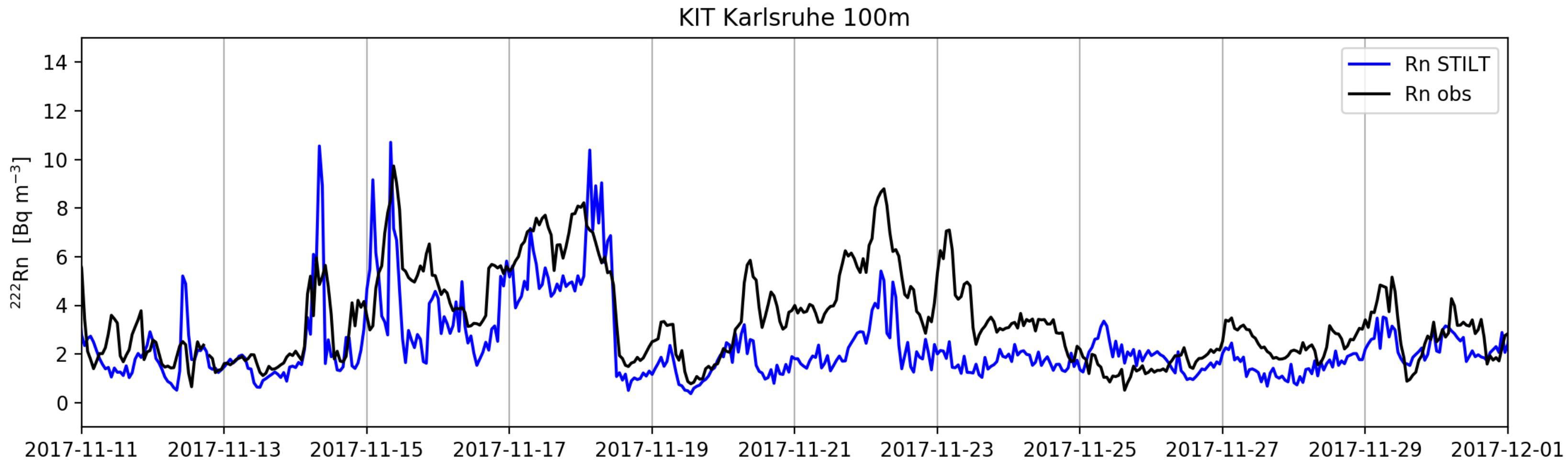
*HRM = Heidelberg Radon Monitor

Winter and summer episodes of atmospheric Radon

Summer:
- pronounced diurnal cycle



Winter:
- variations on synoptic (3-5 days) time scale
- diurnal cycle less pronounced



black line: Observations
blue line: STILT model simulation results using the climatological Radon flux map

Causes of variations on different time scales

- Diurnal cycle
  - Reflects (diurnal) variations of vertical mixing processes in the Atmospheric Boundary Layer: Night-time increase due to accumulation of Radon in the shallow nocturnal boundary layer and decreasing and low values due to strong vertical mixing during the day.
  - The amplitude of the diurnal cycle is also sensitive to the strength of local fluxes (but fluxes have NO diurnal cycle).

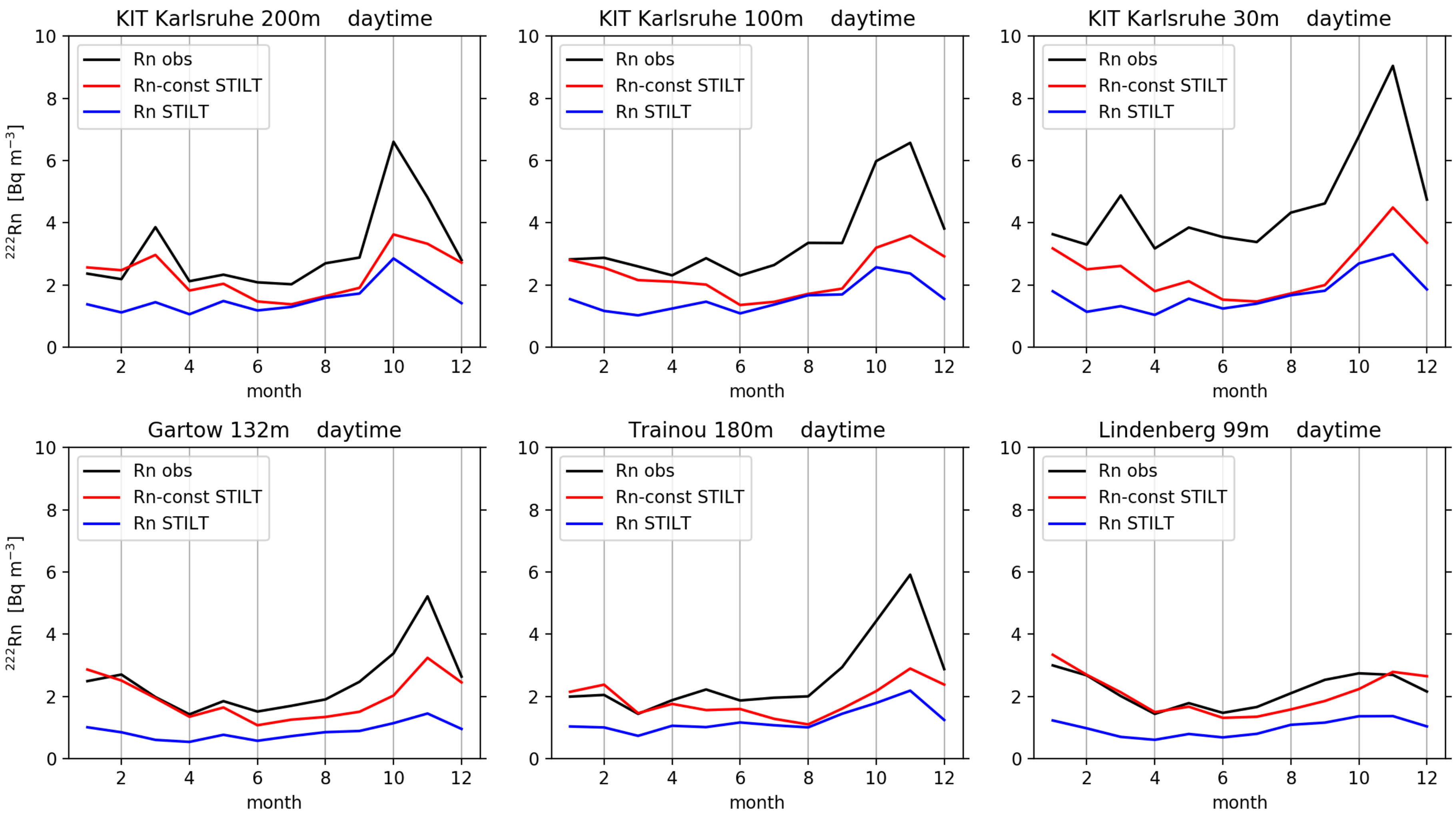
- Synoptic scale variations
  - Are caused by variation in air mass origin, i.e. continental versus marine air and by changes in boundary layer transport conditions.
  - Also regional differences in soil fluxes contribute to synoptic scale variations.

- Seasonal cycle
  - Combined influence of seasonal variations in atmospheric transport patterns and in Radon fluxes as higher soil moisture in winter and early spring reduces the diffusion and exhalation of Radon from soils.

Seasonal cycle of observed and modelled daytime Radon

KIT tall tower measurements at 200, 100, and 30 m

3 tall towers



black line: Observations
blue line: STILT model simulation results using the climatological Radon flux map
red line: STILT model simulation results using a constant Radon flux from land surfaces

daytime (12-16 UTC ≙ 13-17 local time) data was used

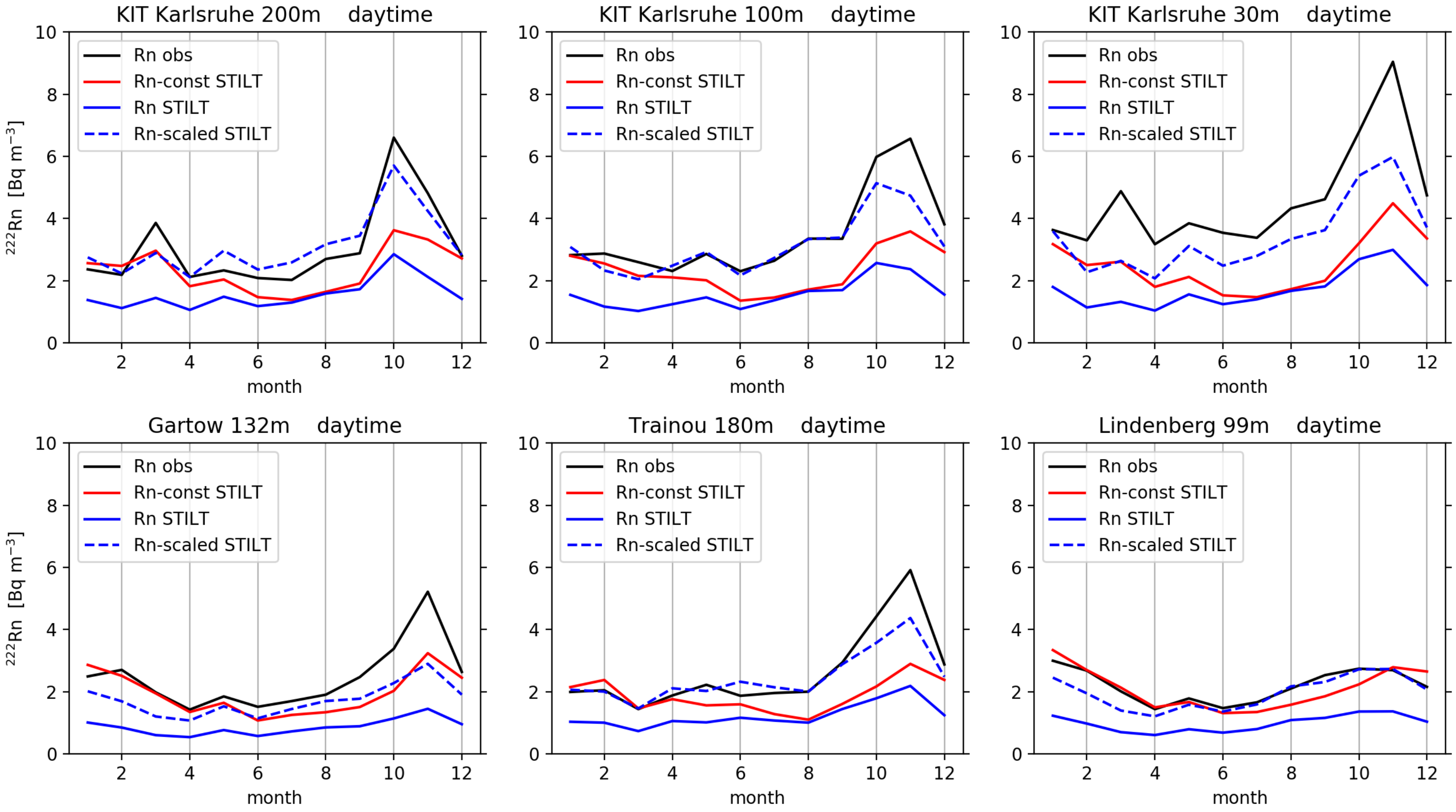
Seasonal cycle of observed and modelled daytime Radon

- Atmospheric transport models are known to have larger uncertainties in representing nocturnal boundary layer processes. Therefore, the seasonal cycles shown in the previous slide are based on daytime values (13h – 17h) only.
- STILT simulations using the climatological Radon flux map, which includes temporal and spatial variability systematically underestimate atmospheric Radon throughout the year.
- STILT simulations using a constant Radon flux show a better agreement with observations in late winter – early spring but a similar underestimation in summer and autumn.
- The shape of the seasonal cycle - in most cases - is better represented using the Radon flux map, which has lower Radon fluxes in winter and spring due to higher soil moisture.
- A simple scaling of simulations with seasonally varying climatological Radon fluxes, as shown in the next slide, results in better agreement between model and observations; this could point to a deficiency in the Radon flux map, i.e. an underestimation of the real flux.

Seasonal cycle of observed and modelled daytime Radon

KIT tall tower measurements at 200, 100, and 30 m

3 tall towers



black line: Observations
blue line: STILT model simulation results using the climatological Radon flux map
dashed blue line: STILT simulations with a *scaled* climatiological Radon flux map
red line: STILT model simulation results using a constant Radon flux from land surfaces

daytime (12-16 UTC ≙ 13-17 local time) data was used

Results from the flux scaling experiment

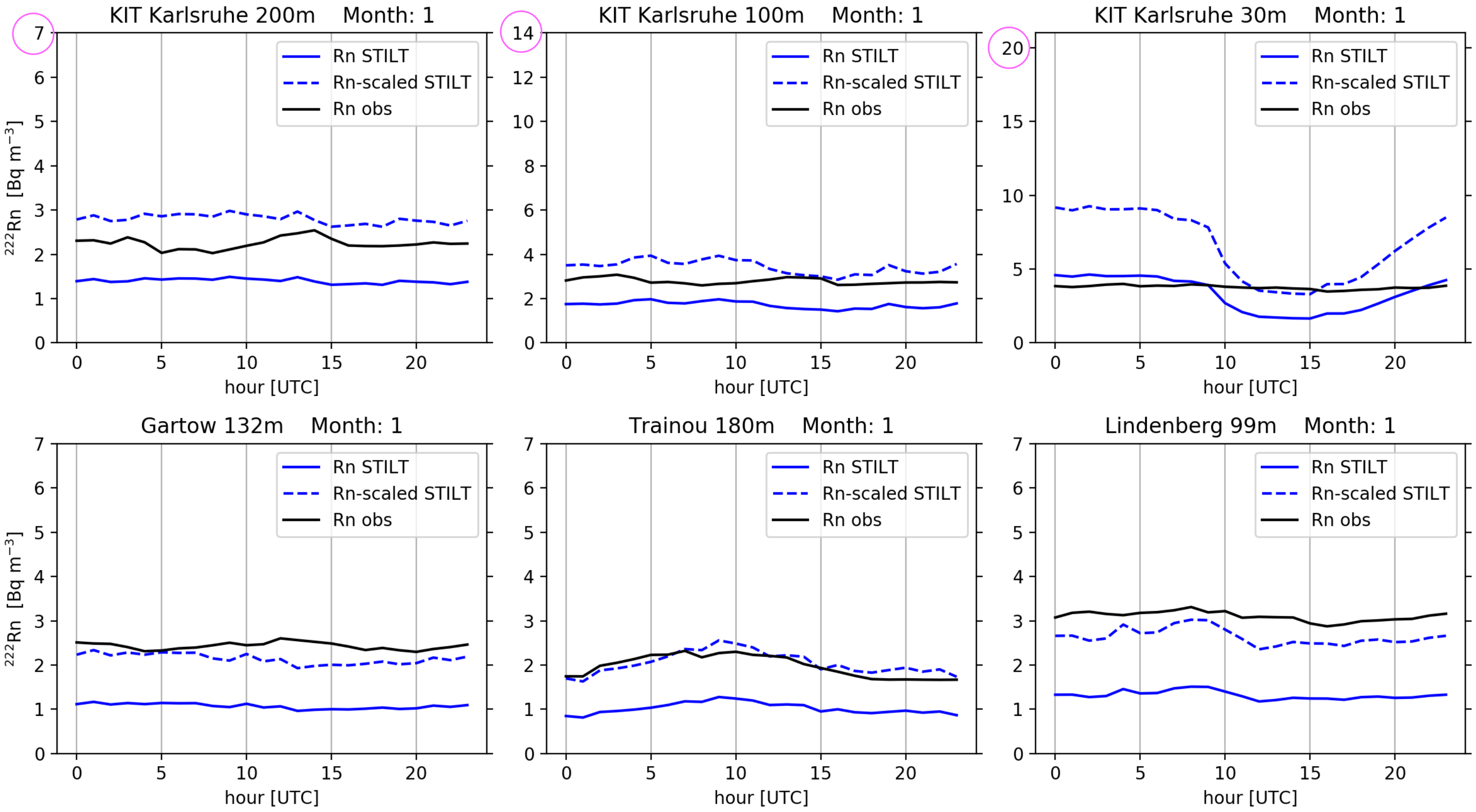
- Doubling of the Radon flux improves the overall agreement between STILT simulations and observed daytime Radon activity concentrations at the high measurement levels (at and above 99 m)

- At the lowest level of KIT tower (30 m), the model still underestimates observations in all seasons.

Diurnal cycle of observed and modelled Radon in winter

KIT tall tower measurements at 200, 100, and 30 m

y-axis scales are different !



3 tall towers

black line: Observations
blue line: STILT model simulation results using the climatological Radon flux map
dashed blue line: STILT simulations with a *scaled* climatiological Radon flux map

Diurnal cycle of observed and modelled Radon in winter

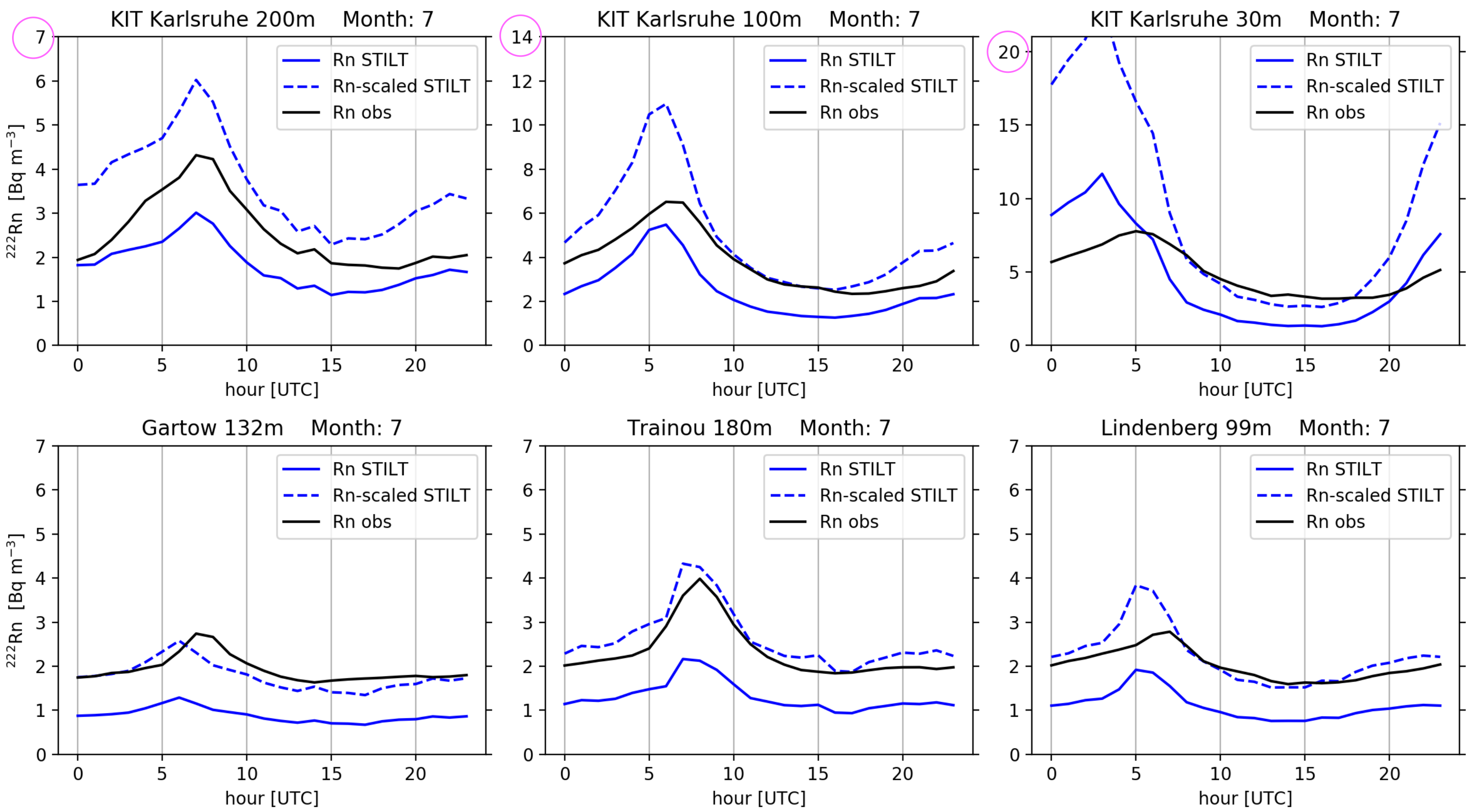
- In winter the observed Radon activity concentration is fairly constant throughout the day.
- At the higher levels of the towers the simple scaling approach results again in a better agreement between modelled and observed Radon at most stations.
- At the lowest level of the KIT tower (30 m) the model shows a clear diurnal cycle that is not present in the observations; doubling of the flux increases the mismatch during night.
- The latter indicates that the model has problems representing vertical mixing in the lower levels of the boundary layer, at least during night time.

Diurnal cycle of observed and modelled Radon in summer

KIT tall tower measurements at 200, 100, and 30 m

y-axis scales are different !

3 tall towers



black line: Observations
blue line: STILT model simulation results using the climatological Radon flux map
dashed blue line: STILT simulations with a *scaled* climatiological Radon flux map

Diurnal cycle of observed and modelled Radon in summer

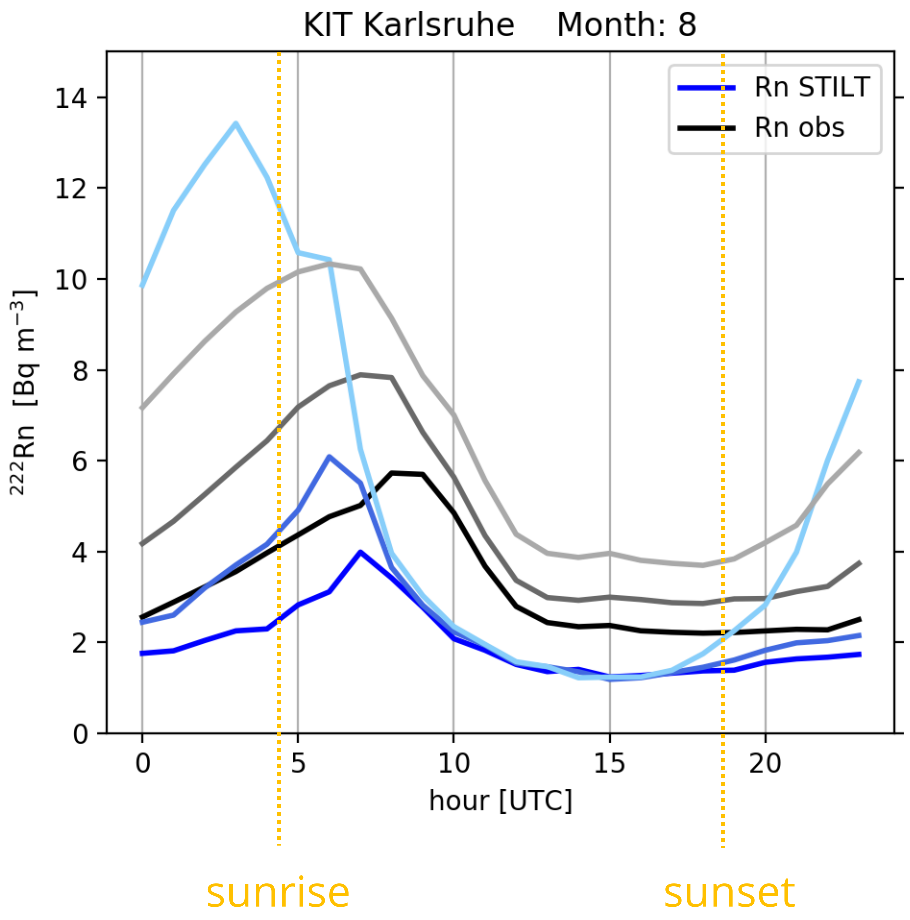
- In summer the diurnal cycle with accumulation of Radon in the nocturnal boundary layer and lower activity concentration during daytime, when the atmosphere is well mixed, is clearly visible in observations and model results.

- The simple flux scaling improves the agreement with observations during daytime at all measurement levels. But it leads to an even stronger overestimation of the model during night time. This is particularly pronounced at the lowest measurement level at the KIT tower (30 m).

- The phase of the diurnal cycle is shifted in the model where the decrease due to vertical mixing starts earlier in the morning compared to the observations. This phase shift increases closer to the surface.

- Again this points to deficiencies in the vertical mixing processes in the model. The vertical gradient measurements at the KIT tower allow a more detailed analysis.

Vertical Radon profile at KIT tower



modeled Rn at **30**, **100** and **200** m

observed Rn at **30**, **100** and **200** m

Vertical Radon profile at KIT tower

- The modelled diurnal cycle is now based on the original climatological Radon flux in oder to focus on potential explanations for the mismatch in the model's process representation.
- In the evening around sunset the concentration close to the surface (30 m) starts to increase quickly in the model, resulting in a very strong vertical gradient with a much larger concentration difference between 30 m and 100 m compared to the observation.
- In the morning the mixing starts 2-3 hours earlier in the model and quickly mixes the accumulated Radon upwards.
- During daytime the vertical gradient in the model disappears due to strong mixing while in the observations a concentration gradient is maintained, also in the afternoon.
- These findings indicate that the vertical mixing in the model is probably too weak during night, resulting in an overestimation of the Radon activity concentration close to the surface producing a strong vertical gradient during night. This behaviour would be even amplified when using higher Radon fluxes.
- On the other hand during daytime mixing in the model is probably too strong. This is indicated by the disappearance of a vertical gradient at KIT.
- Too strong vertical mixing during day in the model could also be the reason for the under-estimation of Radon activity concentration at all stations throughout the year (see comparison of modelled and observed seasonal cycles during the day).

Conclusions

- Our preliminary results have pointed to some important deficiencies in the atmospheric transport model STILT, which are:

- Vertical mixing during night in the lowest boundary layer (30 m) is too much suppressed, leading to too high activity concentrations at that level and too small activity concentrations above. This seems to be the case during summer and winter.

- Vertical mixing during the day seems to start too early in the day then being too vigorous and leading to too low activity concentrations at all levels up to 200 m.

- STILT simulations based on the climatological flux map with higher Radon fluxes during summer than during winter show better agreement in the shape of the seasonal cycle between model and observations than simulations with constant fluxes.

- It can, however, not be ruled out that the climatological flux map generally underestimates real Radon fluxes from soils in Europe.