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# What the “peaks” in local dose rate measured during the Fukushima accident tell about deposition, air concentration and release of radioactivity

GRS

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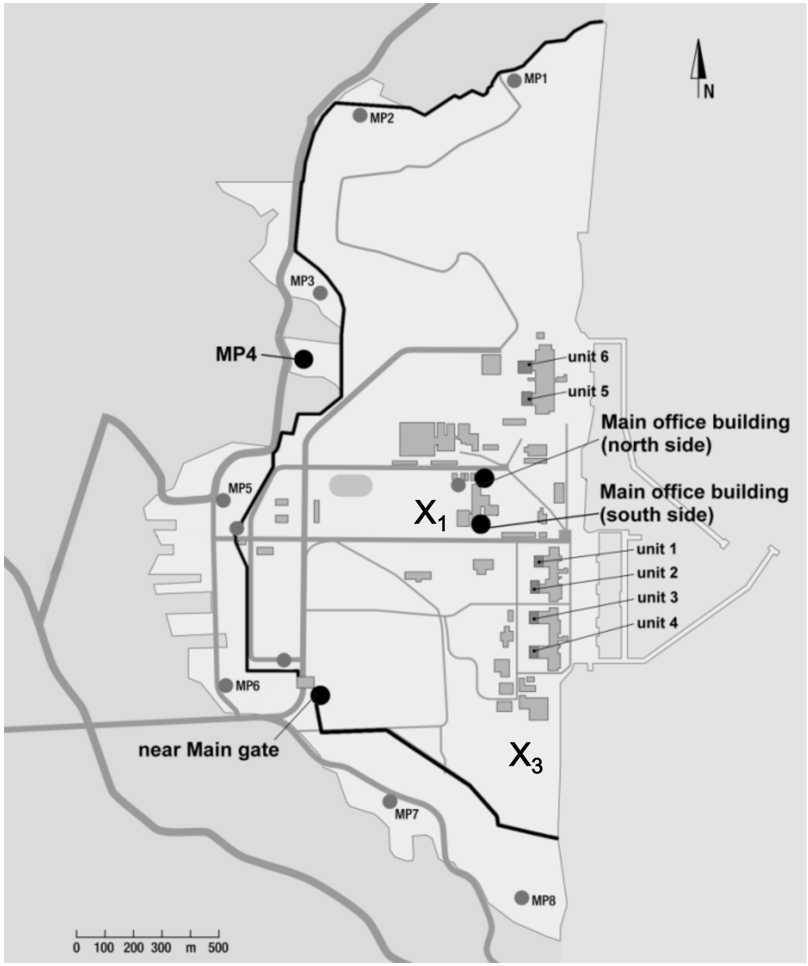
## Scope and objectives

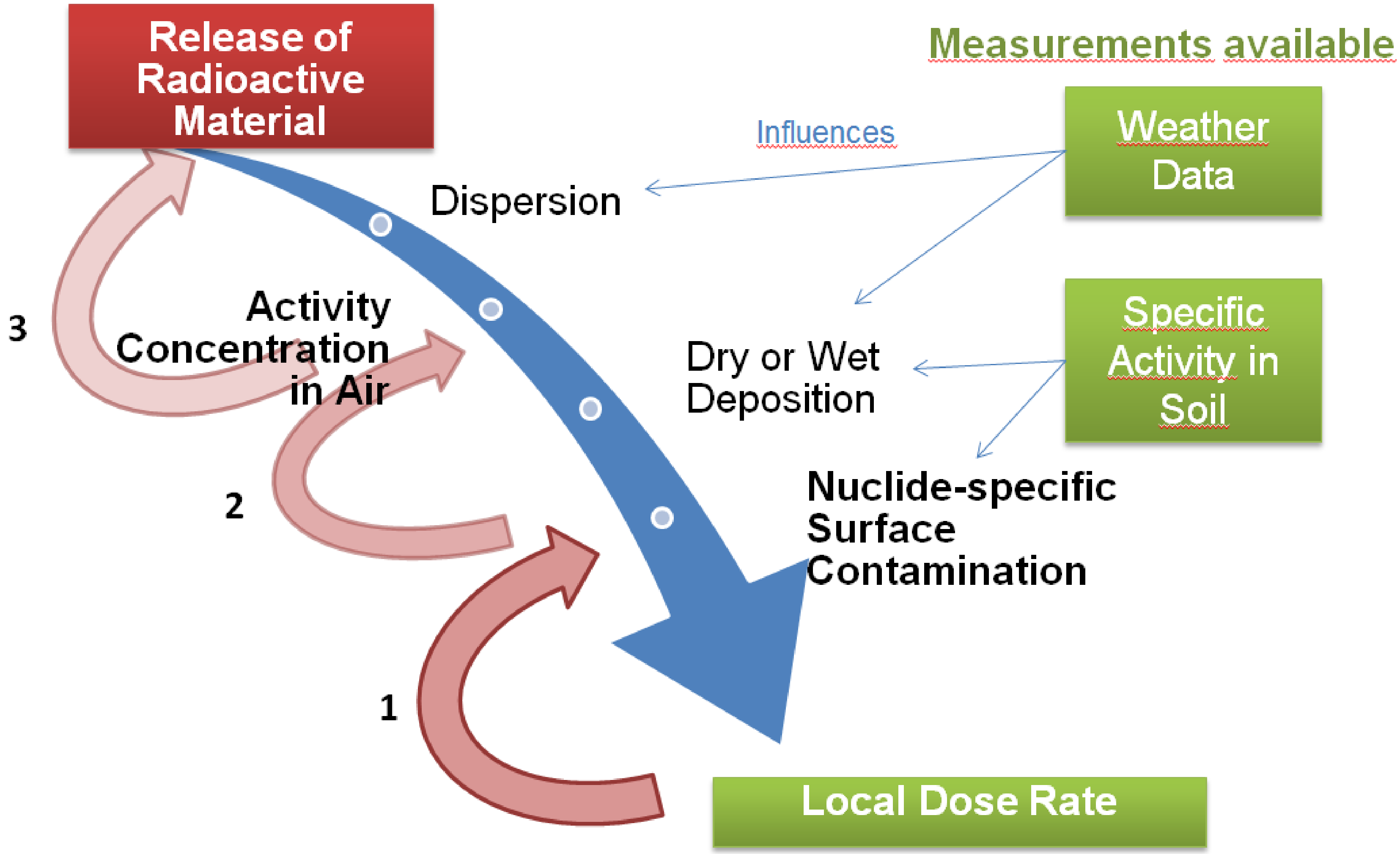
- Investigations are part of OECD/NEA project: "Benchmark Study of the Accident at the Fukushima Daiichi Nuclear Power Plant (BSAF)“, Phase II
  - Severe accident (SA) analyses for Units 2, 3 covering first 3 weeks
  - GRS: Simulations with ATHLET-CD/COCOSYS
- Comparison of radiological evidence with results of SA analyses
  - Reconstruct radioactive releases from measured local dose rate on-site Fukushima NPP or nearby
  - Identify relevant processes for radioactive releases from the plant
  - Draw conclusions on processes and uncertainties which sensitively influence source term estimation
  - Compare results with modelled releases by SA analyses performed within OECD/NEA BSAF Project, Phase II

# Data (1)

- Numerous local dose rate measurements on-site Fukushima Daiichi NPP
- On-site wind, precipitation measurements
- Few soil samples of deposited nuclides (location “$x_1$”, “$x_3$”, from March 21, 2011)
- No measurements of air concentration during release phases
- No measured source term

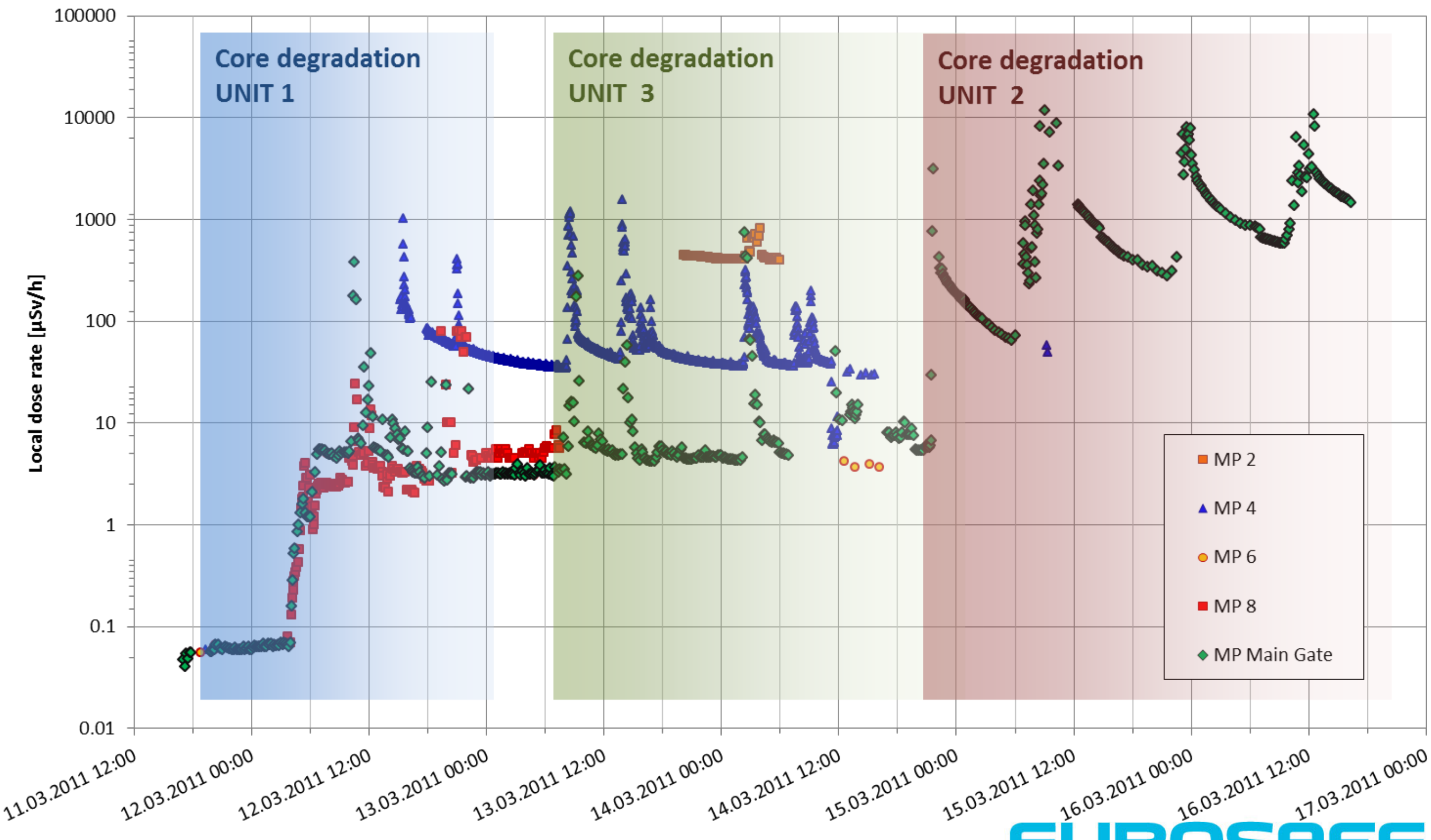


# Reconstruction method for radioactive releases



# Observed local dose rates (LDR) and core degradation in Unit 1-3

- Severe core degradation phases in Units 1 – 3 can be related to measured “peaks”
- Not every “peak” has been linked to events in the plant yet

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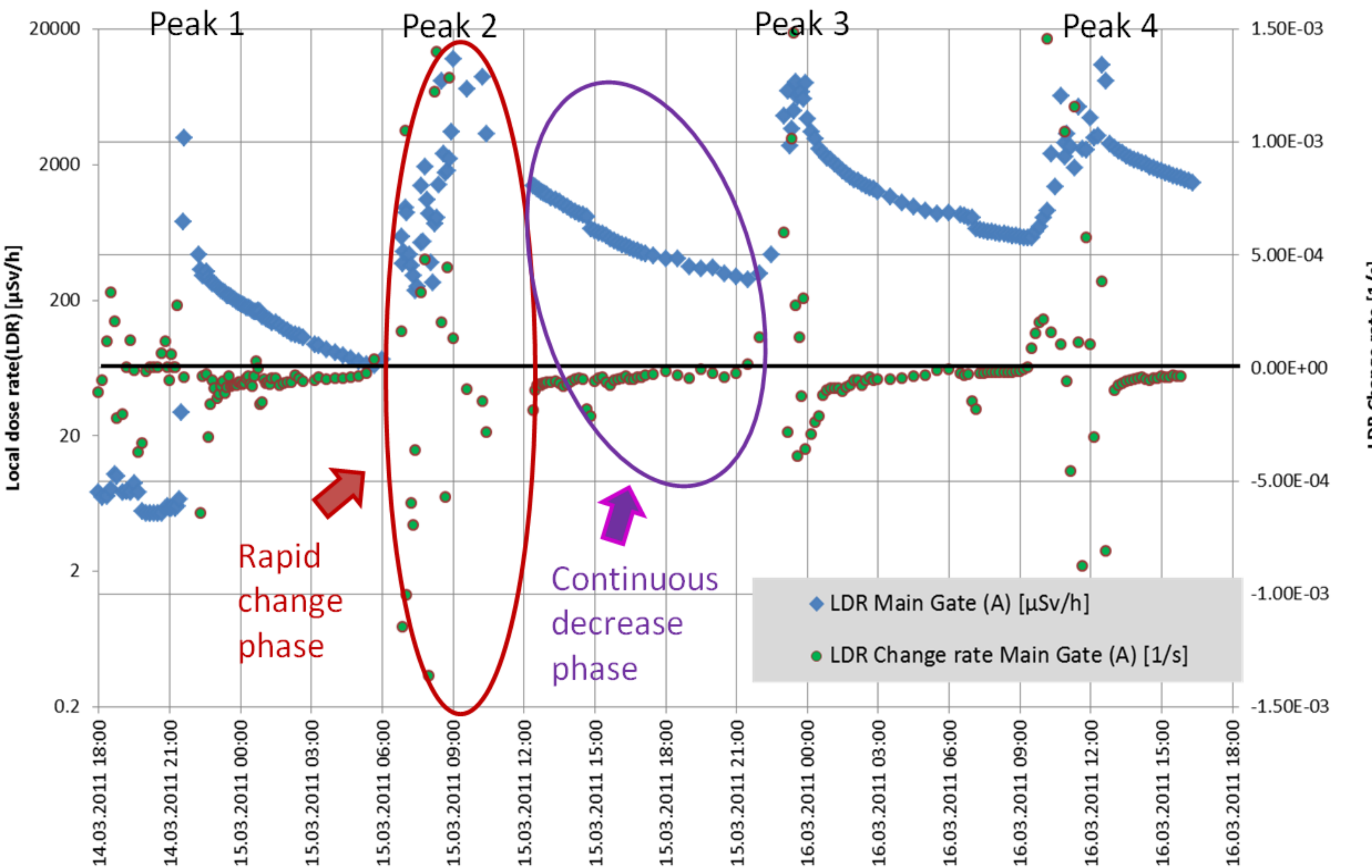
# Structure of largest peaks in local dose rate (LDR)

- The LDR peaks measured at the **main gate** between **March 14, 2011 18:00 JST and March 16, 2011 16:00 JST** have been investigated in detail for this purpose, and are discussed here.
- **Local Dose Rate** $\boldsymbol{\dot{H}}$ and its normalised **Change Rate** $\boldsymbol{\frac{1}{\dot{H}}\frac{d\dot{H}}{dt}}$ are considered.
- All four LDR peaks measured in this time interval exhibit similar structures:
  - **a phase of strong and discontinuous increase and decrease** ("rapid change phase") followed by
  - **a slow and continuous decrease** ("continuous decrease phase")
  - Our objective is to explain observed $\dot{H}$ and $\frac{1}{\dot{H}}\frac{d\dot{H}}{dt}$ in the "continuous decrease phase".

# Structure of largest peaks in local dose rate (LDR)



# Explanation of LDR behaviour in the continuous decrease phases: Test of basic hypothesis

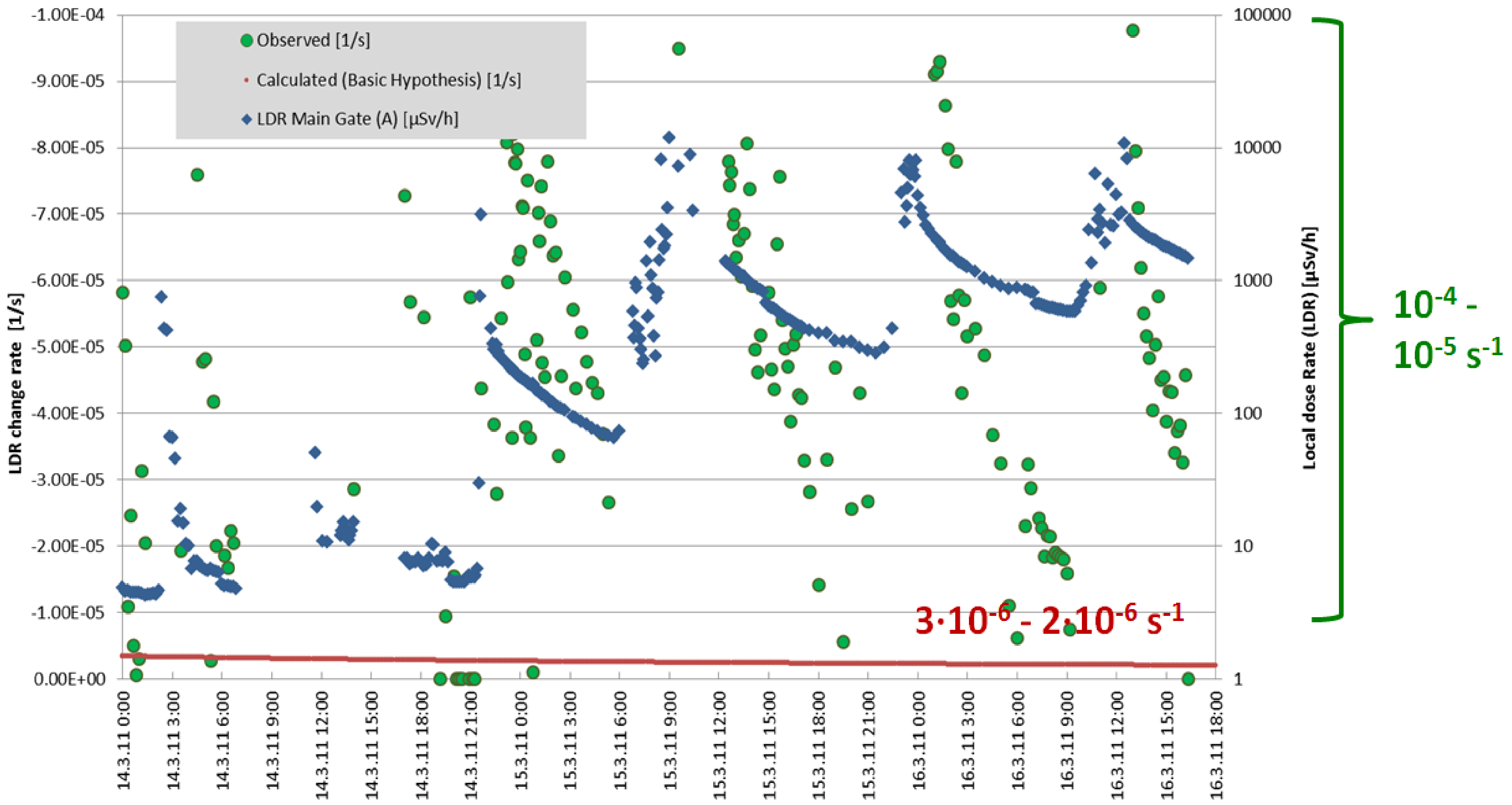
- **Basic hypothesis**: *"The observed LDR is dominated by decay of nuclides deposited on the ground. The nuclide composition can be obtained from soil samples."*
  - **Soil sample from March 21, 2011, decay-corrected for calculation** (see next slide).
  - **I-131 taken from soil measurement.**
  - **Possible contributions from I-132 – I-135** which were produced by fission before scram, taken into consideration as follows:
    - Decayed by March 21, but possible relevant before
    - Calculated from I-131 contamination and respective inventory ratio at scram (decay-corrected)
    - Analysis shows only relevant contribution from I-133.
  - Comparison between observed and calculated change rate in LDR

# Properties of selected nuclides considered in surface contamination

| Nuclide | Dominant Generation Process | Half Life Time | Typical activity inventory of a BWR with same power as Units 2, 3 at Scram [Bq] | Activity concentration in soil sample on March 21, 2011 near playground [Bq/m³] | "Basic Mixture": Activity concentration in hypoth. soil sample on March 15, 2011 00:00 JST with equal composition [Bq/m³] |
|---|---|---|---|---|---|
| I-131 | Fission | 8.02 d | 1.9 E+18 | 5.80 E+06 | 9.74 E+06 |
| I-132 | Fission | 2.3 h | 2.8 E+18 | in Equilibrium with Te-132 | <<1 |
| | Decay of Te-132 | | | | in Equilibrium with Te-132 |
| I-133 | Fission | 20.7 h | 3.8 E+18 | n/a | 1.83 E+06 |
| I-134 | Fission | 52.5 min | 4.3 E+18 | n/a | <<1 |
| I-135 | Fission | 6.63 h | 3.7 E+17 | n/a | 5.42 E+03 |
| Ru-106 | Fission | 1.005 yrs. | 1.5 E+18 | 5.30 E+04 | 5.36 E+04 |
| Te-129m | Fission | 33.6 d | 7.0 E+16 | 2.50 E+05 | 2.83 E+05 |
| Te-132 | Fission | 3.18 d | 2.7 E+18 | 6.10 E+05 | 2.25 E+06 |
| Cs-134 | Fission | 1.998 yrs. | 3.4 E+17 | 3.40 E+05 | 3.42 E+05 |
| Cs-136 | Fission | 13.15 d | 1.2 E+17 | 7.20 E+04 | 9.88 E+04 |
| Cs-137 | Fission | 30.108 yrs. | 2.4 E+17 | 3.40 E+05 | 3.40 E+05 |
| Ba-140 | Fission | 12.73 d | 3.2 E+18 | 1.30 E+04 | 1.80 E+04 |
| La-140 | Fission | 1.67 d | 3.2 E+18 | 3.30 E+04 | 3.93 E+05 |

Calculated from I-131 soil activity concentration and inventory ratio at scram

# Observed local dose rate in the continuous decrease phase vs. basic hypothesis




**Calculated LDR decrease rate magnitude is much too low**

$(3 \cdot 10^{-6}$ - $2 \cdot 10^{-6}$ $s^{-1}$)compared to $10^{-4}$ - $10^{-5}$ $s^{-1}$ observed LDR decrease rate

# Explanation of LDR behaviour in the continuous decrease phase: Test of alternative hypotheses (I)

**Alternative hypothesis 1**: *“The observed LDR is caused by the radioactive cloud which is slowly drifting away and disperging.“*

- Lower limits for characteristic time scales in LDR caused by very slow drift or diffusion of radioactive clouds calculated with dispersion model ARTM

⇒ Cloud drift: > $10^{-3}$ $s^{-1}$; Cloud diffusion > $10^{-4}$ $s^{-1}$

Moving and disperging cloud causes change rates that fit the „rapid change phase“ but not the „slow decrease phase“ (with $10^{-4}$ - $10^{-5}$ $s^{-1}$).

**Alternative hypothesis 2**: “*The observed LDR is caused by reduced and slowly decreasing radioactive releases after a strong release from the reactor(s) (puff release + continuous release).“*

In that case, the LDR signal at the measuring point would also be modulated by atmospheric dispersion, with change rates caused by the drifting and disperging cloud in contradiction to observations during the continuous decrease phase.

# Explanation of LDR behaviour in the continuous decrease phase: Test of alternative hypotheses (II)

**Alternative hypothesis 3**: *“The observed LDR is caused by deposited radio- nuclides which are slowly reduced by wind-driven resuspension and/or runoff by rainfall.“*

- Resuspension by turbulence: timescales shorter than $10^{-4}$ $s^{-1}$.
- Precipitation has been only recorded after March 15, 22:30 JST

Resuspension or runoff cannot produce the systematic behaviour during all four continuous decrease phases after the largest peaks

⇒ **None of alternative hypotheses 1 - 3 can explain observed LDR development in the slow decrease phase.**

# Explanation of LDR behaviour in the continuous decrease phase: Test of alternative hypotheses (III)

**Alternative hypothesis 4**: *"The observed LDR is caused by deposited radionuclides **including short-lived nuclides** that must have been produced considerable time after scram and shortly before release."*

**Potential production processes:**

a. **Excess release of short-lived daughters of fission products** which are more volatile than their mother nuclides (in particular I-132 as a daughter of Te-132)

- Efficiency of process unknown
- Assuming the same release fraction for I-132 as for I-131, an upper bound for excess release of I-132 compared to Te-132 can be guessed from measured I-131 and Te-132 contamination and inventories in the core:

⇒ Calculated average ratio **I-132 : Te-132** ≅ **4 : 1**

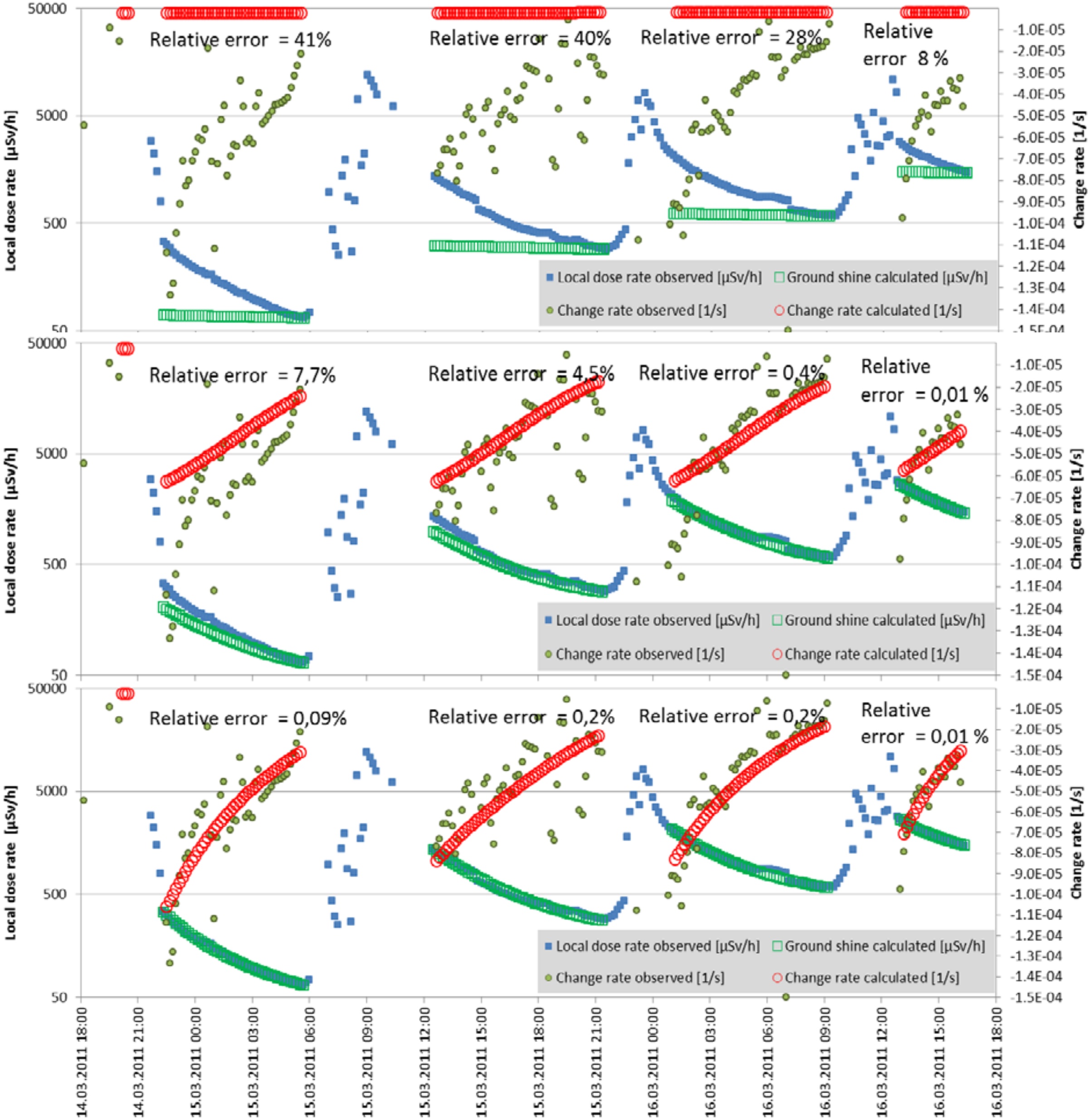
⇒ Estimated upper bound for contamination ratio **I-132 : Te-132** ≅ **8 : 1**

b. **Additional fission products release from recriticality events** in particular I-134, I-132 in core.

# Explanation of LDR behaviour in the continuous decrease phase: Test of hypothesis 4

- Hypothesis 4 is tested by simulating ground shine with different assumptions on composition of deposited nuclides
  - Simulation A: Composition as in the „basic mixture“
  - Simulation B: Additional contamination by I-132 ($t_{1/2}$ = 2 h 20 min ) as volatile daughter of Te-132 (constrained by a 8:1 ratio to Te-132)
  - Simulation C: Additional contamination by I-134 ($t_{1/2}$ = 52,5 min ) and I-132 from potential recriticality events
- The optimum composition of nuclides is estimated by Monte Carlo simulation for each continuous decrease phase.
- The agreement between calculated and observed LDR is compared for the three simulations to find out which optimally fits the observed development in LDR

# Observations and simulated nuclide mixtures




**Simulation A:**
„Basic Mixture“

**Simulation B:**
Excess release of I-132 as daughter of Te-132 with a maximum ratio 8:1

**Simulation C:**
Additional release of I-134 and I-132 from potential recriticality event

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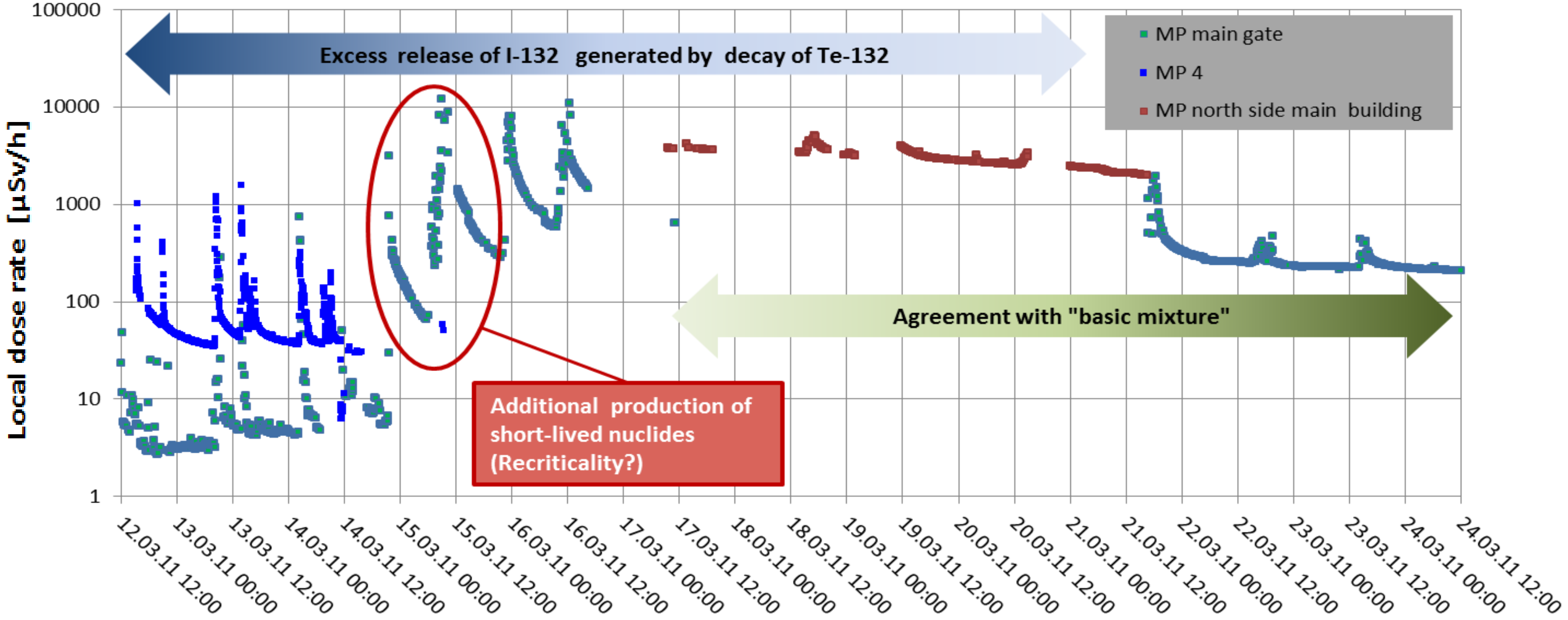
# Conclusions from analysis of peaks 1 - 4

- ***Only ground shine with contributions by short-lived nuclides can explain observations in "continuous decrease phases"***
  - These nuclides must have been produced significantly later after reactor scram occurred and are not evident in later soil samples
  - Alternative hypotheses addressing atmospheric or other release processes have been found incapable to explain observations
- ***This contribution can be partly attributed to excess release of I-132 which is produced in the core by decay of Te-132.***
  - This leads to higher I-132 activities compared to Te-132 after end of deposition and a subsequent faster decrease in LDR.
- ***For peaks 1 and 2, another production process of short-lived nuclides is needed to explain observed LDR curves***
  - Excess release of I-132 from Te-132 is not efficient enough and contribution by a nuclide with even shorter half-time than I-132 is likely
  - Contributions from I-134 and I-132 from potential recriticality suitably explain observations

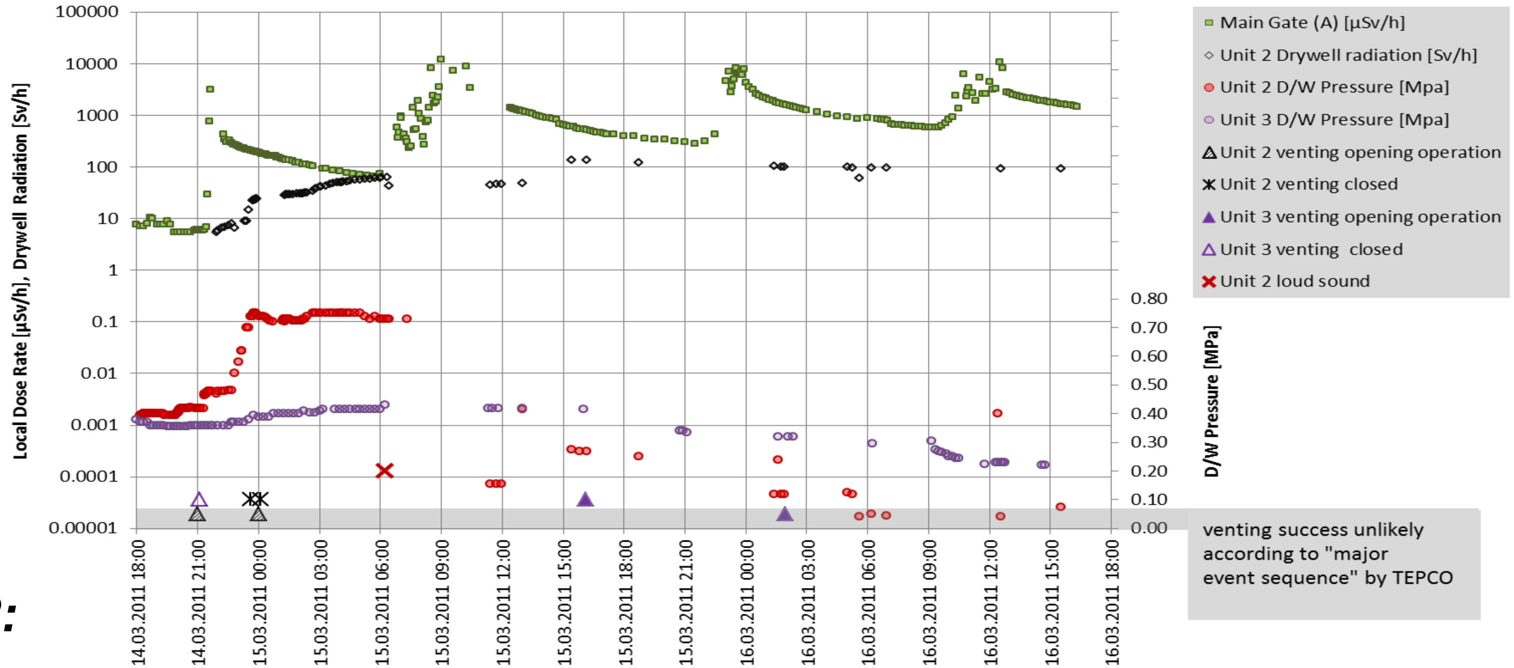
# Comparison with other measuring points and times

- In the first days,  all other continuous decrease phases can be explained by excess release of I-132 from Te-132 decay
  - (calculated ratios  I-132 :Te-132 between 1,4 and 3,5)
- Later phases: Increasingly good agreement with „basic hypothesis“




***Indications of recriticality unique for peaks on March 14 and 15.***

# Comparison with plant parameters




- ***Unit 2:***
  - ***Containment pressure:*** Temporary halt in increase corresponds with peak 1; strong pressure drop during peak 2 “rapid change phase”
  - ***Containment radiation:*** Continuous increase between peak 1 and 2, indicating progressive extension of core damage in this phase
  - ***Reactor water level (not shown):*** Drop below core bottom level before peak 1 followed by intermittent rise of water level, core temporally and partially covered when peaks 1 and 2 occur – recriticality?
- ***Unit 3:*** No notable correspondence of peaks 1-4 with plant information

# Discussion and outlook

- ***Analysis of local dose rate peaks and corresponding reactor data indicate possibility for recriticality events in Unit 2 between March 14 late afternoon and March 15 around noon***
- ***Occurrence of recriticality needs to be clarified for analysis of fission product release as it changes nuclide composition***
  - Neglecting contributions from short-lived nuclides will lead to an overestimation of I-131 and Cs-137 releases
- ***Clarification is also essential for explanation of core degradation and observed reactor pressure vessel (RPV) pressure pikes***
  - Do current results of severe accident (SA) analyses agree well with measured containment pressure and RPV pressure?
  - Could “extra energy” by recriticality improve agreement of SA analyses with observed behaviour or will it rather lead to contradictory results?
- ***Assessment of potential occurrence, duration and intensity of recriticality events by severe accident analysis will be very helpful***

**Thank you  for your attention!**