

**IRSN**

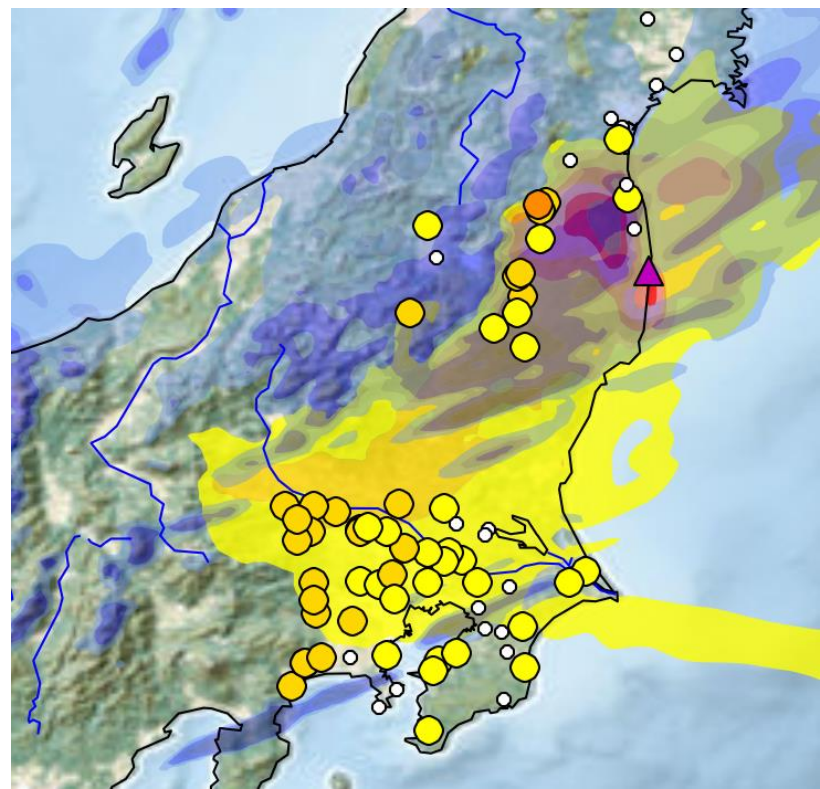
INSTITUT  
DE RADIOPROTECTION  
ET DE SÛRETÉ NUCLÉAIRE

*Faire avancer la sûreté nucléaire*

A new perspective on the Fukushima releases  
brought by newly available  $^{137}\text{Cs}$  air concentration  
observations and reliable meteorological fields

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Work done by  
O. Saunier, A. Mathieu, T. Sekiyama,  
M. Kajino, K. Adachi, M. Bocquet, T. Maki,  
Y. Higarashi and D. Didier



## Role of IRSN in case of a Radiological Emergency

Assess risk induced by accidental situation - Provide technical expertise to public Authorities

Before Fukushima the IRSN method to assess a ST was based on the analysis of the state of the power plant only (has to be done by facility expert).

+ Essential to forecast the emissions. Fully independent from the observations in the environment and from errors due to ATM, Met data...

When too few information from the plant are available, the approach is useless.

- Provide a rough estimation of the ST.

Past events proved that it can be tricky to precisely estimate the releases in the atmosphere and the consequences

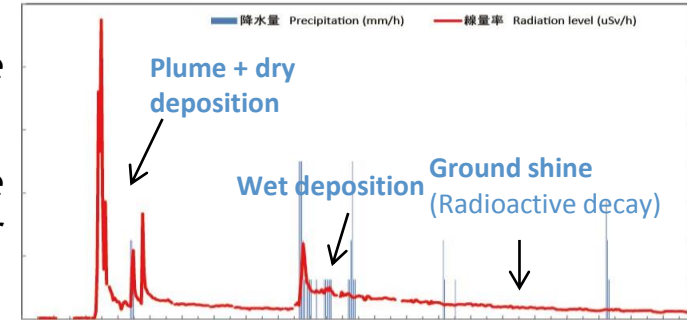
Main nuclear accidents (Fukushima) and minor events (radionuclide detection by monitoring system: iodine detection 2011-2012, cesium detection 2013 and 2015, forest fires in areas contaminated by the Chernobyl accident...).

➤ Need to develop a complementary operational method to assess accurately the emissions by using environmental observations.

(ST: temporal evolution of the release rate + distribution between radionuclides)

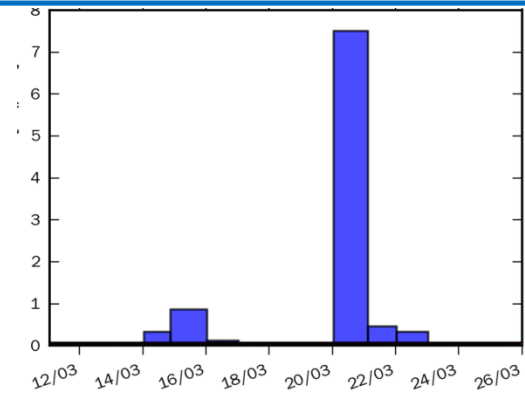
## Gamma dose rate measurements

- ☺ High temporal resolution, dense spatial coverage, available in real time.
- ☹ No access to the isotopic composition of the ST and to the respective share of the plume and the deposit. Available for major release events only (high detection level).



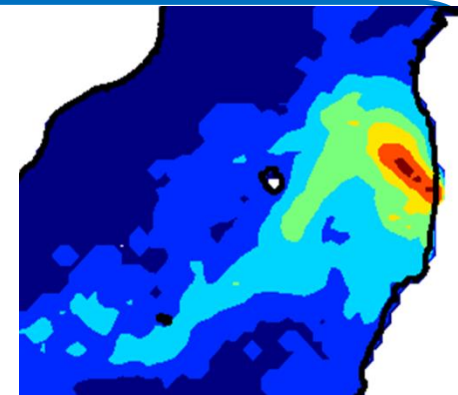
## Air concentration & daily deposition measurements

- ☺ Provide information on the isotopic composition of the release. Available for major + minor release events: low detection level.
- ☹ Coarse spatial coverage. Time averaged data (often 24 h). Time series not always available. Delays in making data available.



## Total deposit

- ☺ Dense spatial coverage. Provide information on the isotopic composition of the release.
- ☹ No information on the temporal evolution of the deposit during the release period. Delays in making data available.



## □ Source-receptor relationship

$\mu$	=	$H$		$\sigma$	+	$\epsilon$
Vector of observations		Source receptor matrix <i>computed with the forward ATM (Abida et al. 2011)</i>		Estimator of the ST		Vector of errors <i>Observations Model</i>

## □ Variational method

$$J(\sigma) = \frac{1}{2}(\mu - H\sigma)^T R^{-1}(\mu - H\sigma) + \frac{1}{2}(\sigma - \sigma_b)^T B^{-1}(\sigma - \sigma_b)$$

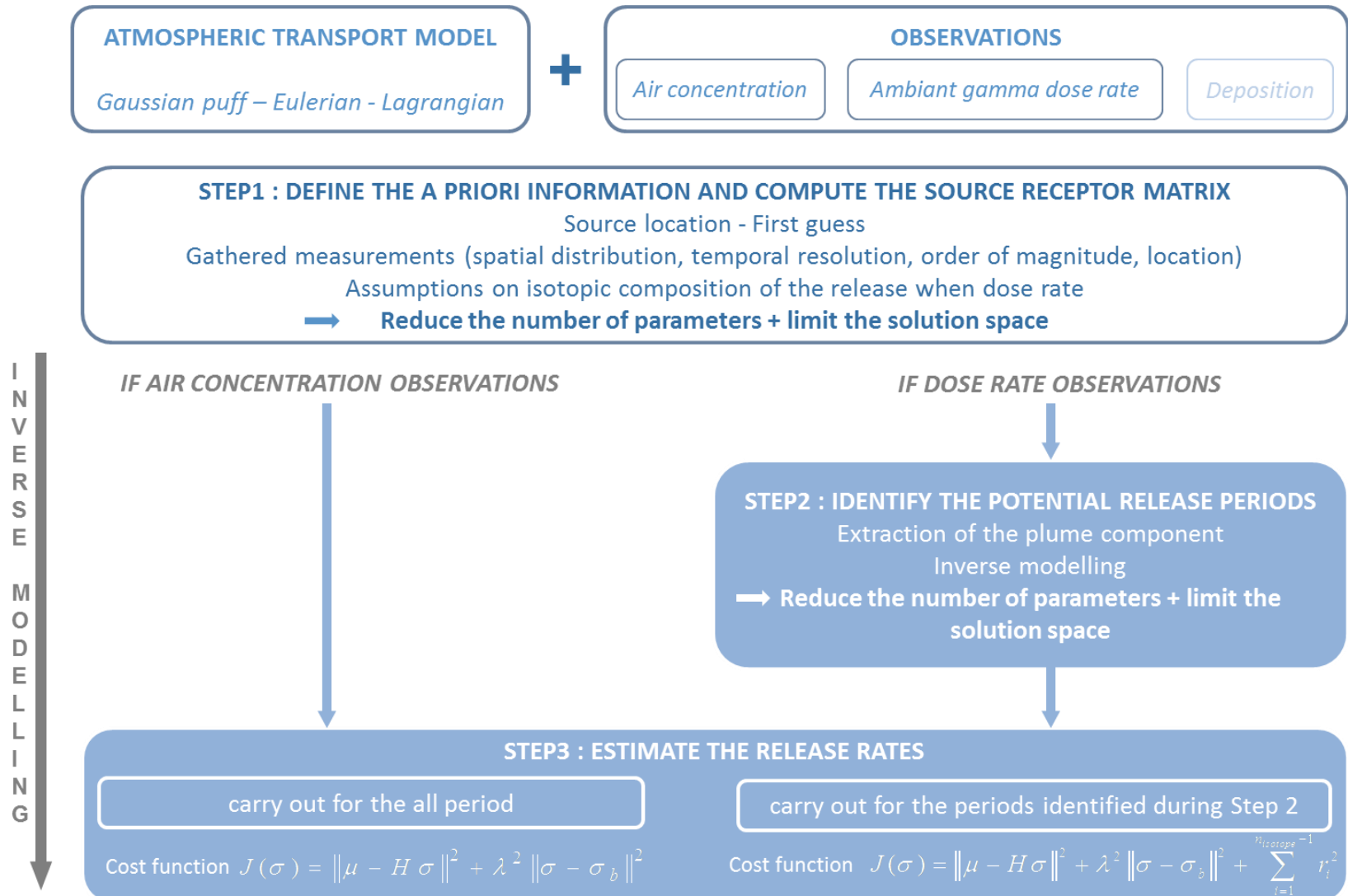
- Cost function  $J$  which measures differences between:
  - Observations and model.
  - a priori and a posteriori emissions.
- Minimisation of  $J$  using gradient descent algorithm (L-BFGS-B method).

## □ Requirements

- Location of the potential accidental sites.
- Number of observations  $\gg$  number of unknowns parameters if *a priori*  $\sigma_b = 0$ .

# Operational tool to assess the Source Term by using observations in the environment

based on inverse modelling approaches (mathematical rigorous approach)



Since 2011 Many source term estimates have been published.

Most of them were estimated by using measurements in the environment

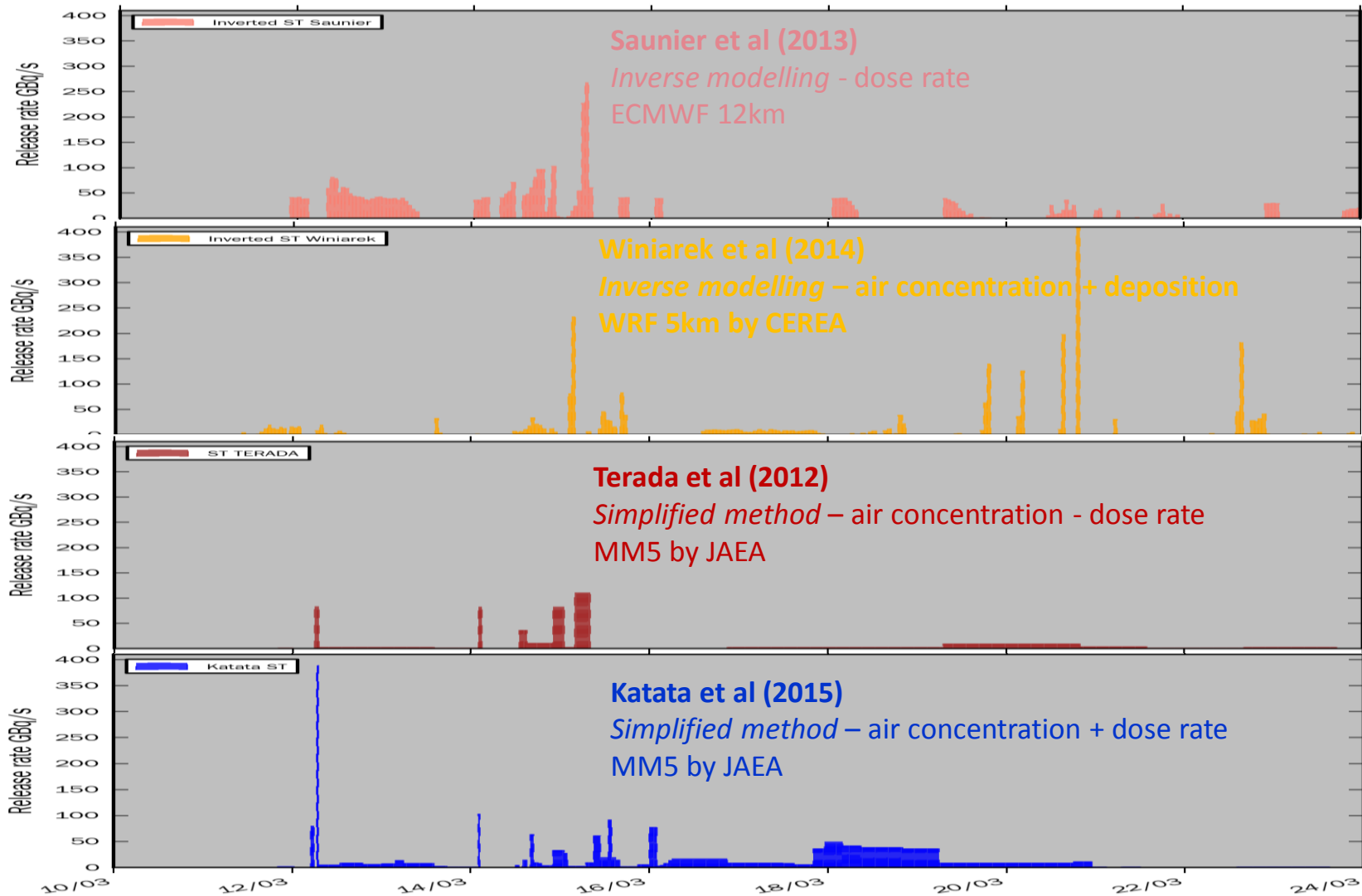
Similar order of magnitude of the total amount released toward the Japanese territory

Source term	Total amount of $^{137}\text{Cs}$ (Pbq)		Method	Observations and A priori
	Japan + Ocean	Japan territories		
NISA (2011)	15		Facility analysis	No temporal evolution
NSC (2011)	12		Facility analysis	No temporal evolution
Chino et al. (2011)	13	7,2	Simplified	Air concentration + dose rate measured in Japan + analysis of the facility events
Stohl et al. (2012)	34,9	23,5	Inverse Modelling	Air concentration measured in N hemisphere + analysis of the facility events
Winiarek et al. (2012)	19	12	Inverse Modelling	Air concentration measured in Japan, US and Canada
Mathieu et al. (2012)	20,6	12,6	Simplified	Air concentration + dose rate measured in Japan + analysis of the facility events
Terada et al (2012)	8,8	5,5	Simplified	Air concentration + dose rate measured close to the FDNPP + analysis of the facility events
Saunier et al. (2013)	15,5	12,3	Inverse Modelling	Dose rate measured in Japan
Winiarek et al. (2014)	11,6-19,3	7,4	Inverse Modelling	Air concentration + deposit measured
Katata et al. (2015)	12,4	8,7	Simplified	Air concentration+ dose rate measured close to the FDNPP and in the ocean + analysis of the facility events
IRSN 2016 not published		7,8	Inverse Modelling	Air Concentration in Japan ( with Tsuruta et al, 2014 obs.)

Same release events

BUT significant discrepancies in the temporal evolution of the release rates ( $^{137}\text{Cs}$ )

for a same release event the amount released can be significantly different



# Is one of those source terms more realistic than the others?

Identify if one of the ST is closer to what really was released in the atmosphere during the Fukushima accident.

## ■ Forward atmospheric dispersion simulations using the various ST as model inputs were carried out.

Simulations were done using the same ATDM (IdX from the C3X platform) with the same configuration.

MRI met. data (Sekiyama et al 2013) was used (3 km).

## ■ Model to data comparison

“New” air concentrations measurements (Tsuruta *et al* 2014, and Oura et al. 2015)

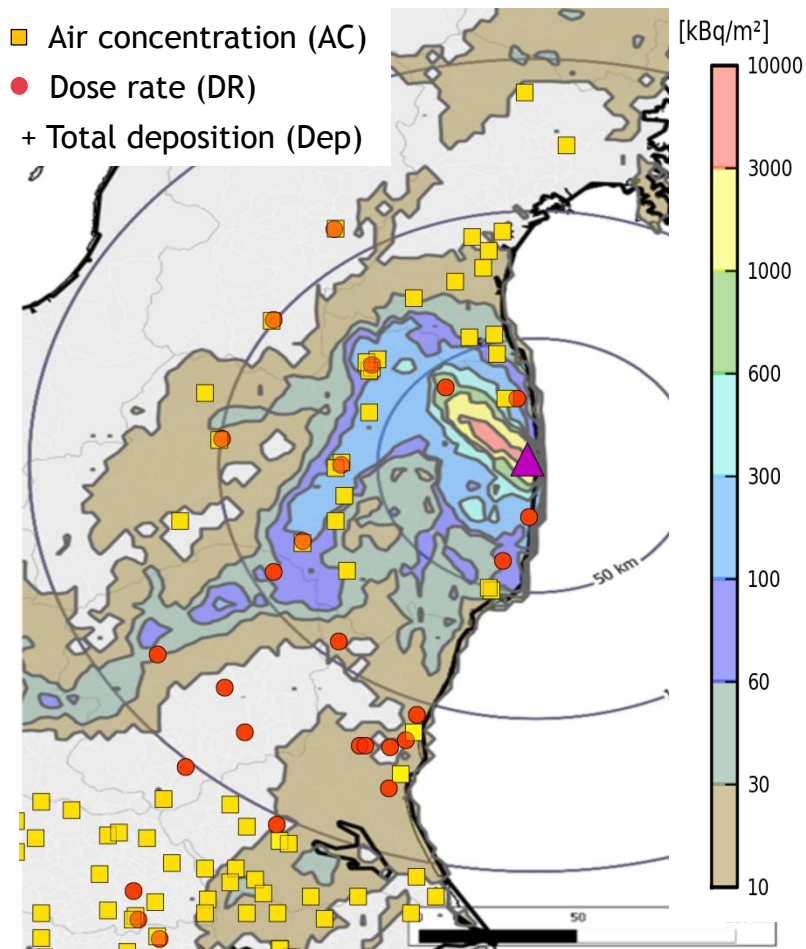
-> ***New point of view***



# Model to data comparison with the Tsuruta et al (2014) observations

Air Pollution Surveillance Network provided  $^{137}\text{Cs}$  air concentration measurements with an hourly temporal time step (11520 obs.).

Data available during 4 periods of time: March 12-13, 15-16, 18-19, 20-23



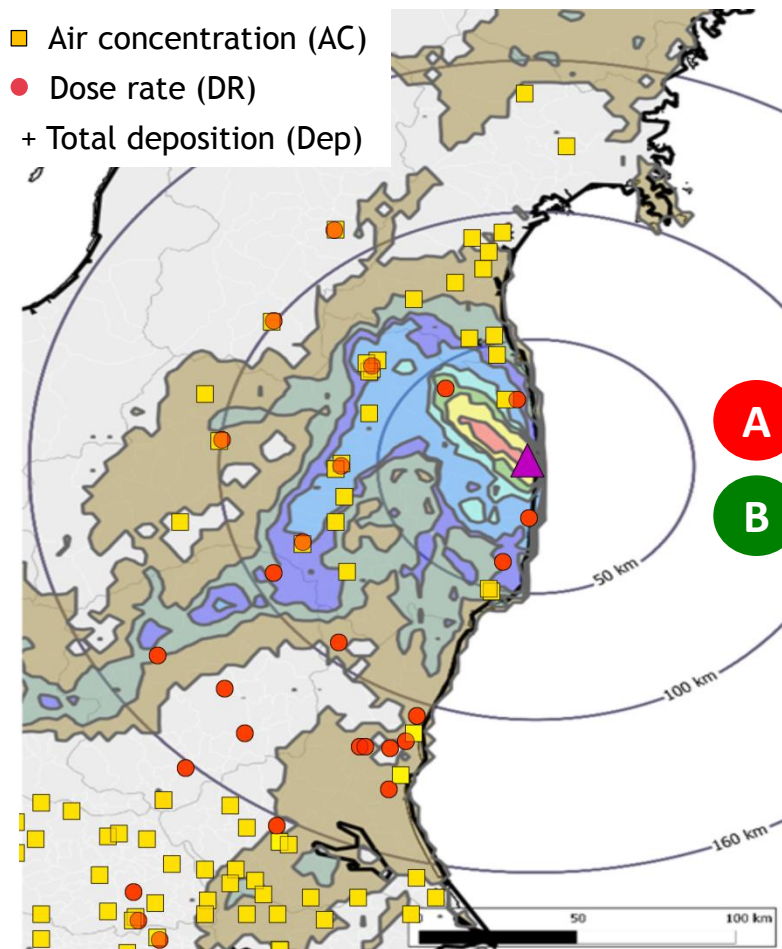
	FAC5(%)
Saunier <i>et al</i> 2013 (DR)	40
Terada <i>et al</i> 2012 (AC, DR)	34
Winiarek <i>et al</i> 2014 (AC, Dep)	37
Katata <i>et al</i> 2015 (AC, DR)	34
Inverted ST (DR + MRI)	37

➔ Model to data comparison shows a weak agreement

# Model to data comparison with the Tsuruta et al (2014) observations

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Data available during 4 periods of time: March 12-13, 15-16, 18-19, 20-23

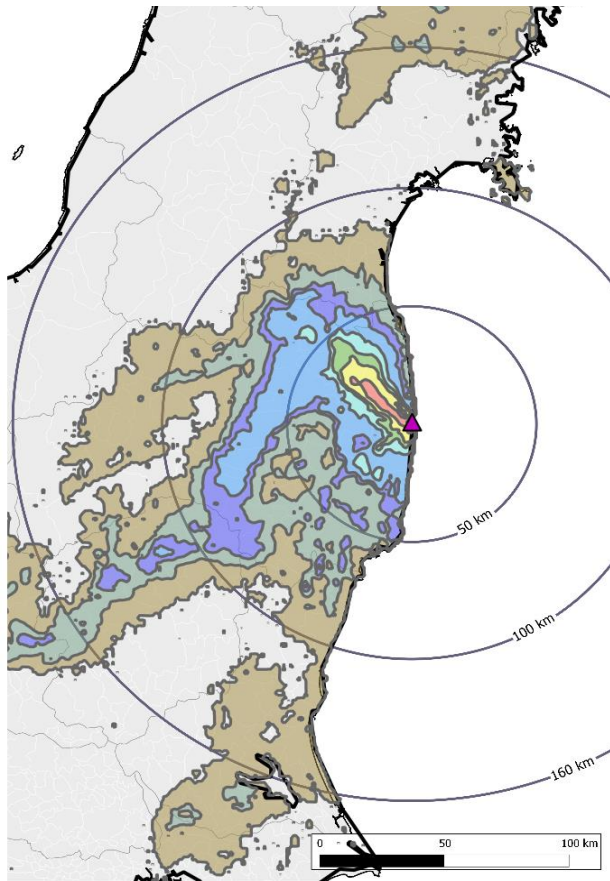


	FAC5(%)
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Katata <i>et al</i> 2015 (AC, DR)	34
Inverted ST (DR + MRI)	37
Inverted ST (Tsuruta AC + MRI)	51

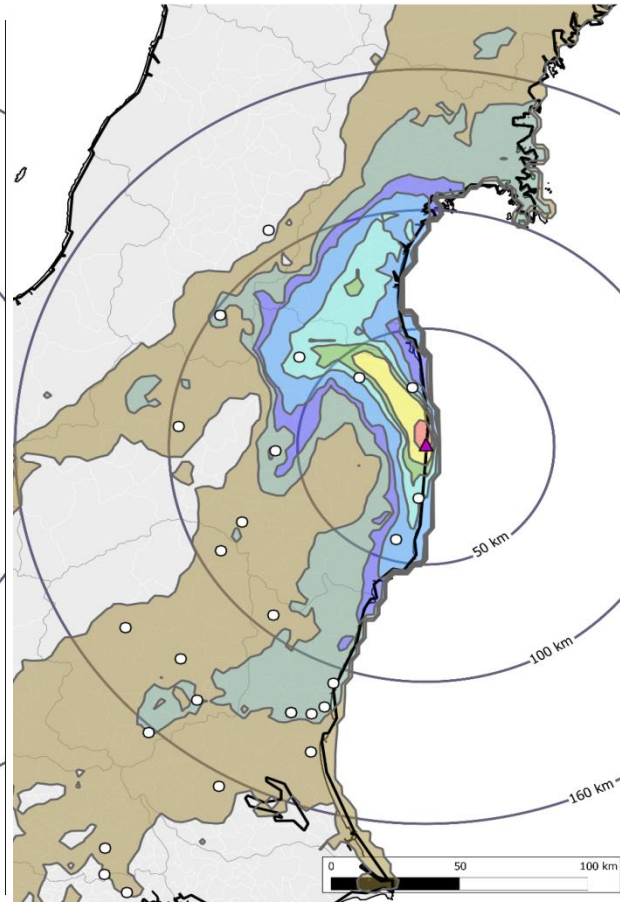
➔ Better agreement with a ST assessed by using Tsuruta et al air concentration measurements

# Are the Tsuruta et al AC measurements the awaited solution for the assessment of the Fukushima ST ?

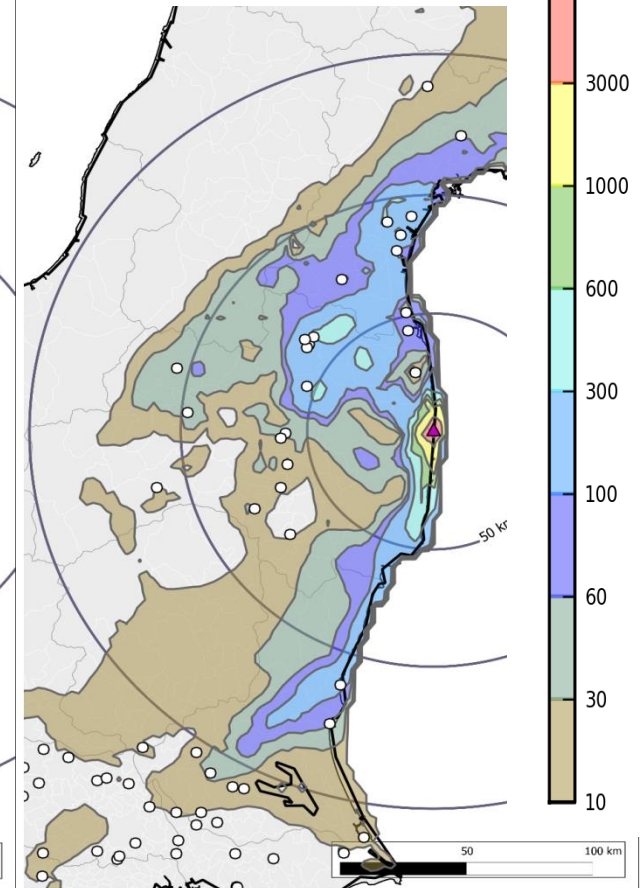
Observed Total  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  deposit  
(US DOE – MEXT 2011)



Simulation with inverted dose rate ST

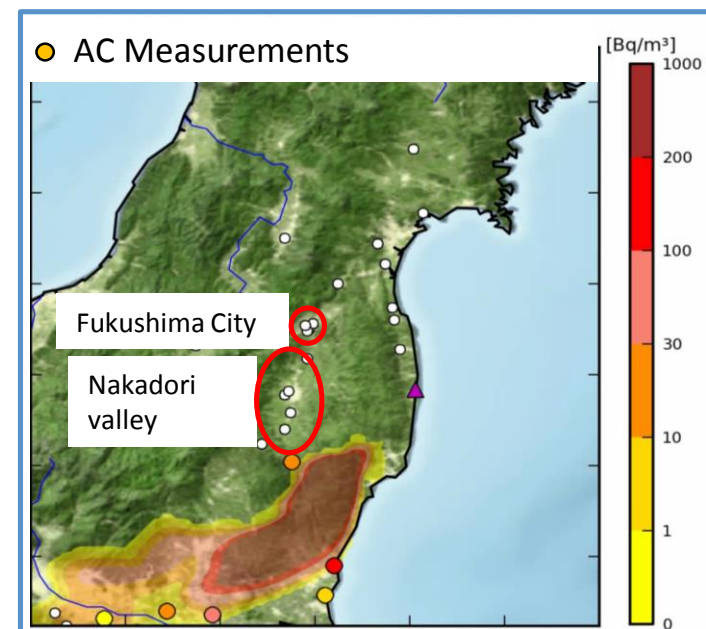
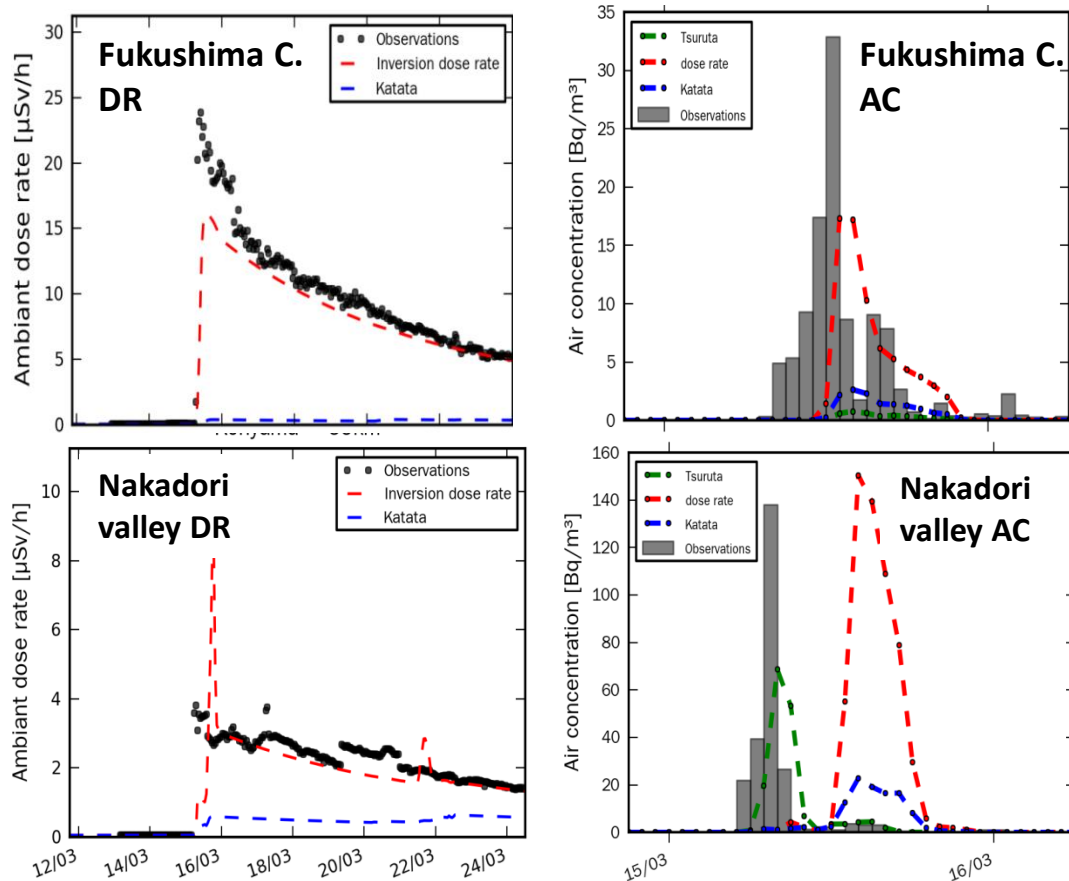


Simulation with inverted Tsuruta et al AC ST [kBq/m<sup>2</sup>]



**A** Simulated deposit (dose rate ST) is more realistic than the one provided by **B** (Tsuruta et al air concentration ST).

## March 15 03:00 –18:00 UTC release event



--- A Simulation done with the inverted ST assessed with Dose Rate.

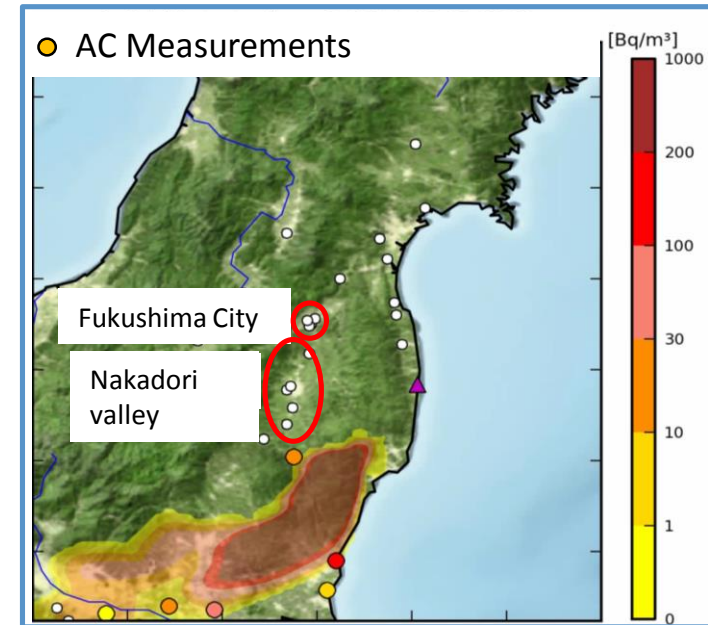
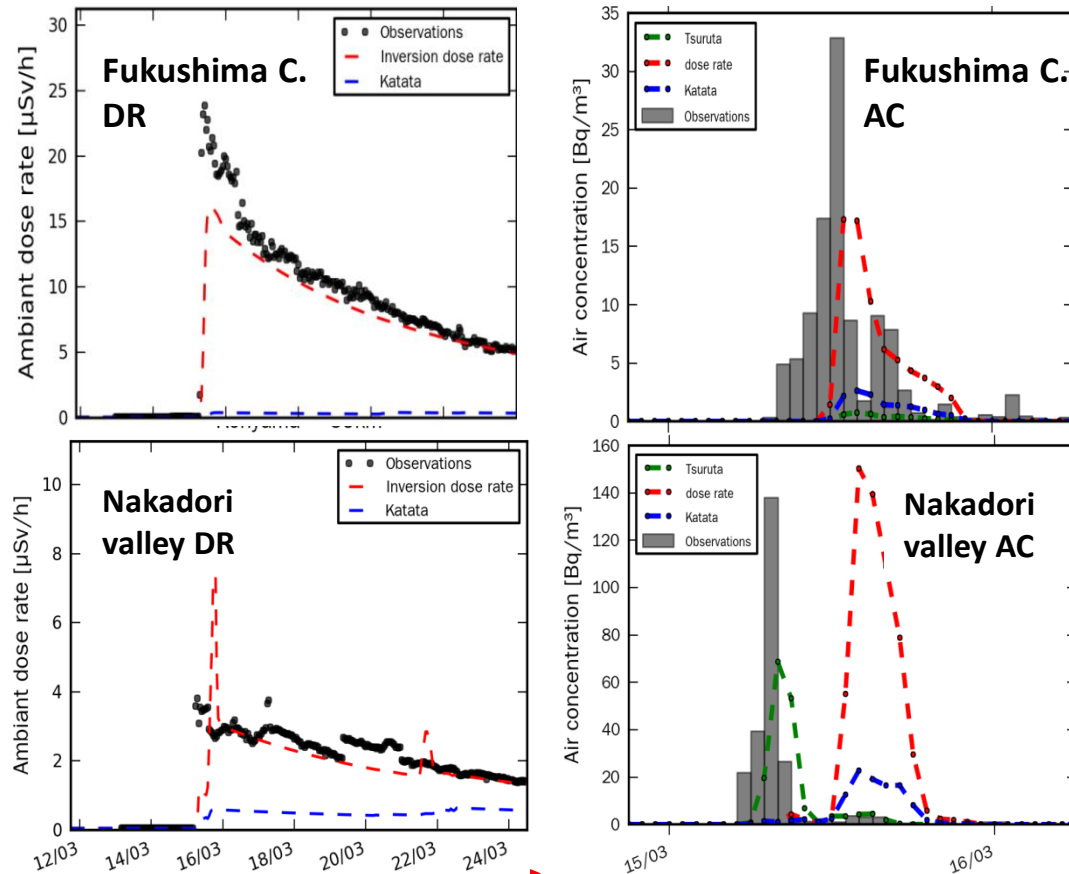
--- B Simulation done with the inverted ST assessed with Tsuruta Air Concentration.

--- Simulation done with Katata et al, 2015 ST

### ➔ Nakadori valley

- Good agreement between **A** simulation and DR but plume is strongly overestimated.
- **B** simulation agrees correctly with observed AC but underestimates the deposit.

## March 15 03:00 –18:00 UTC release event



- **A** Simulation done with the inverted ST assessed with Dose Rate meas.
- **B** Simulation done with the inverted ST assessed with Tsuruta Air Concentration meas.
- Simulation done with Katata et al, 2015 ST

- Inverted dose rate ST (**A**) is designed to fit the observed dose rate signal mainly induced by the contribution of the deposit.
- Inverted air concentration ST (**B**) designed to fit the air concentration observed close to the ground.
- We look for a ST that fit both the deposit and the air concentration.

# Conclusion and perspectives

## Reliable inversed modelling method to assess the source term

- ❑ Very helpful to better understand the Fukushima contamination events.

## Improvements

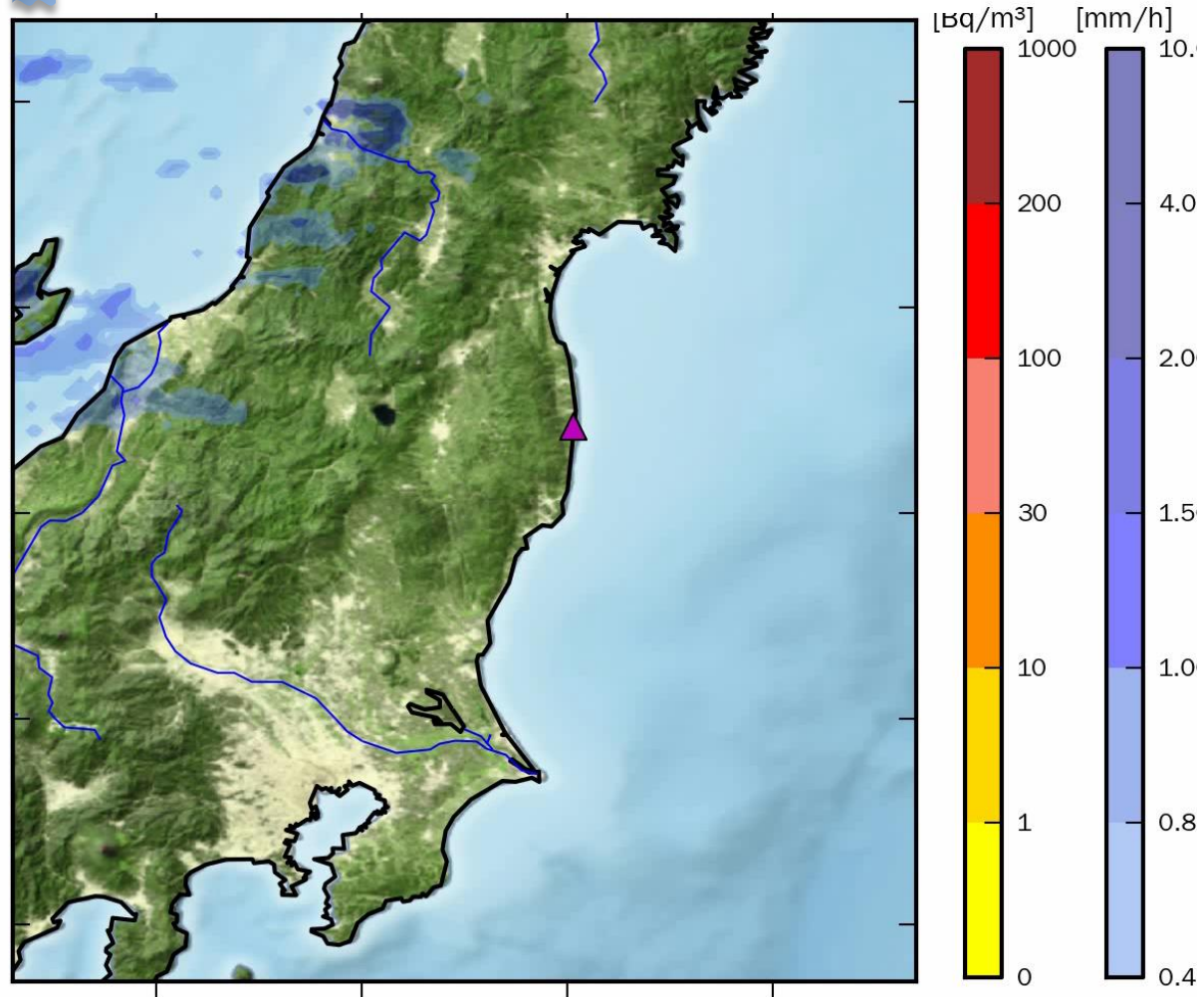
- ❑ Take into account all together different types of observations.
- ❑ Improve the estimation of errors (R and B matrixes).
- ❑ Use a more realistic deposition scheme.

## But this will not be sufficient without more reliable meteorological fields

- ❑ Precipitation fields are a crucial input (begining of the rain events).

Thank you for  
your  
attention!

- Air concentration measurements
- **B** Plume simulation with inverted air concentration ST
- Radar rain observations



# Perspectives

## Reliable inversed modelling method to assess the source term

- ❑ Has to take into account all together the different kind of observations.
- ❑ Has to improve the estimation of errors.
- ❑ Has to use a more realistic deposition scheme.

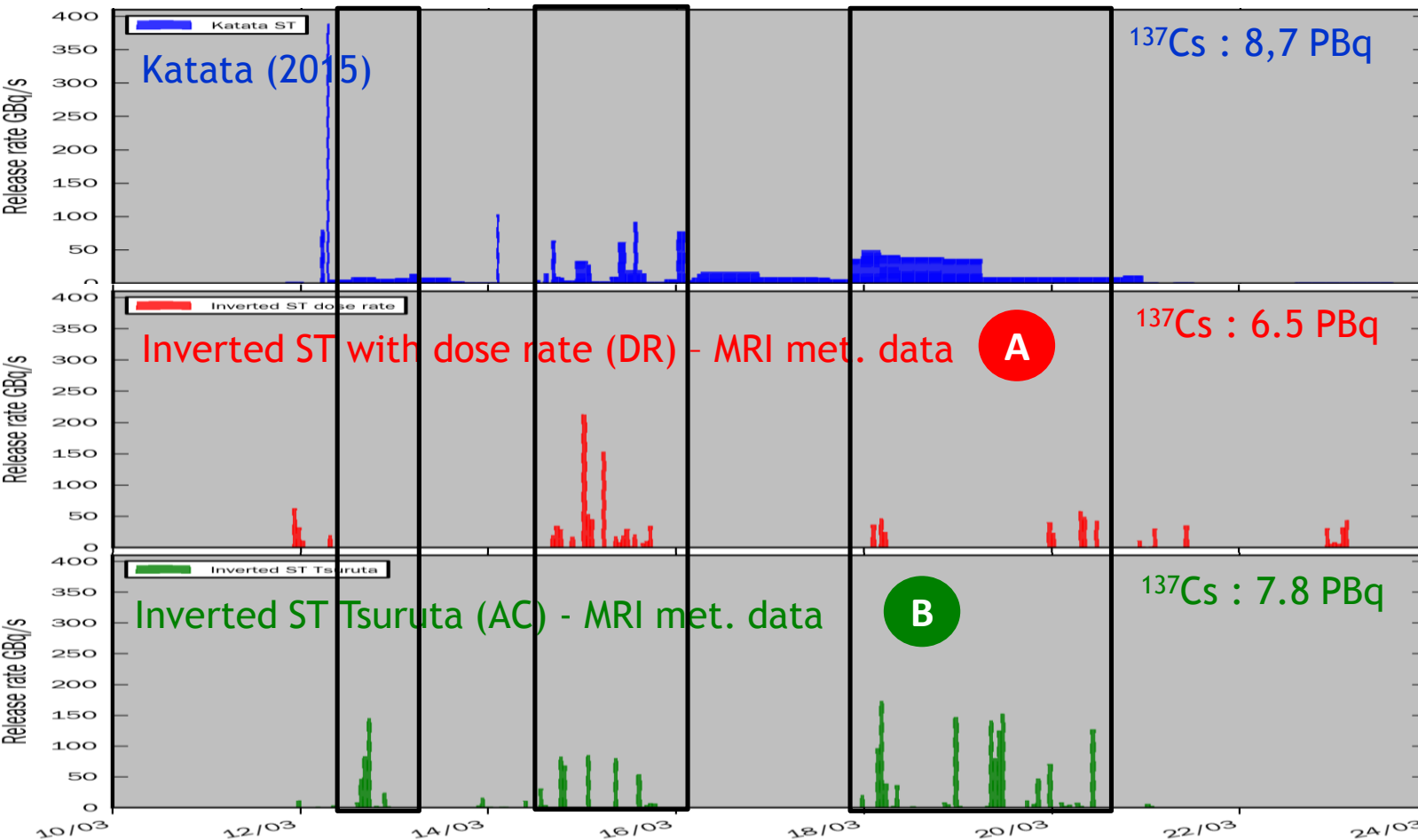
## But this will not be sufficient without more reliable meteorological fields

- ❑ Precipitation fields are a crucial input (beginning of the rain events).

## Other issues

- ❑ Deposition modelling and vertical distribution modelling of the plume.



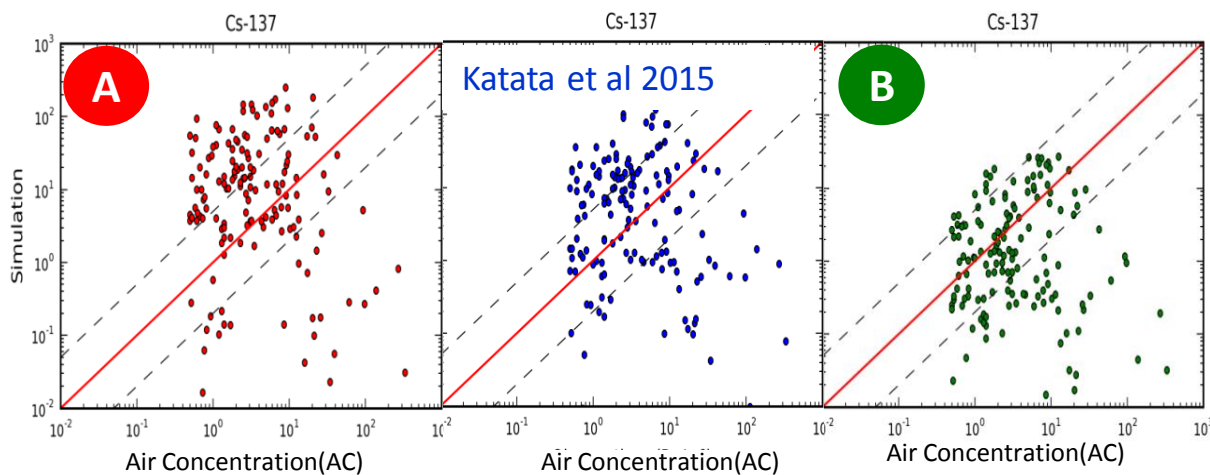


Significant dust releases in reconstituting the event responsible of the main contaminated area located in the NW of the FDNPP

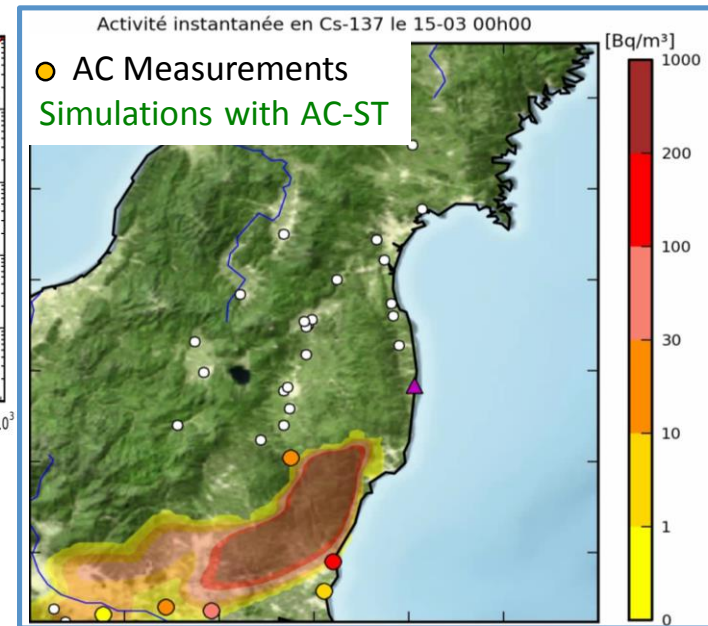
- On March 12 (plume travels northward) and then toward the Pacific ocean ~ not observed with DR
- On March 14-16 (plume travels south westward then westward and north-westward then westward again and southward)
- On March 19 (plume travels southward)
- On March 20-21 (plume travels north westward then southward)

➤ How can we distinguish again and southward between A and B ?

Example : March 15 03:00 –18:00 UTC



	<b>A</b>	Katata et al 2015	<b>B</b>
FA5	30	40	54



➤ Overestimation of the AC by simulating the event with the **A** al ST.

➤ Better agreement obtained for simulations done with **B** ST.

**A** Simulations done with DR-ST: (inverted ST assessed with Dose Rate meas. + MRI)

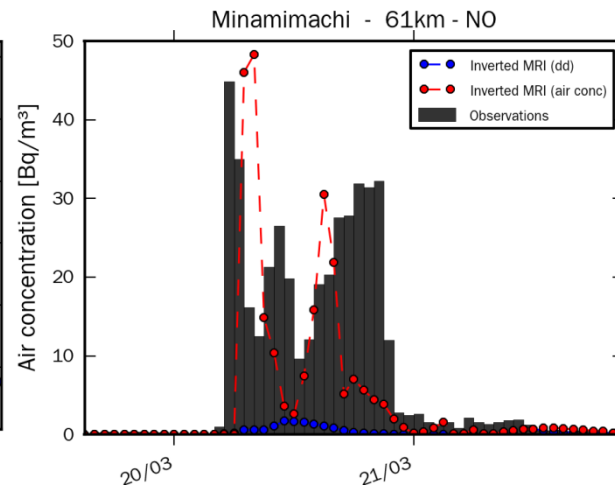
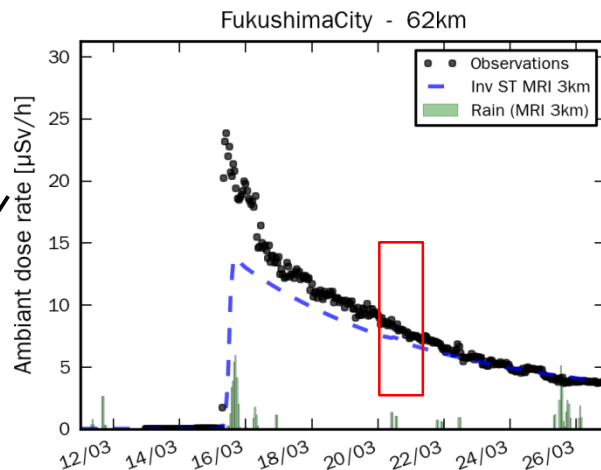
**B** Simulations done with AC-ST : (inverted ST assessed with Tsuruta Air Concentration meas. + MRI)

Simulations done with Katata et al, 2015 ST

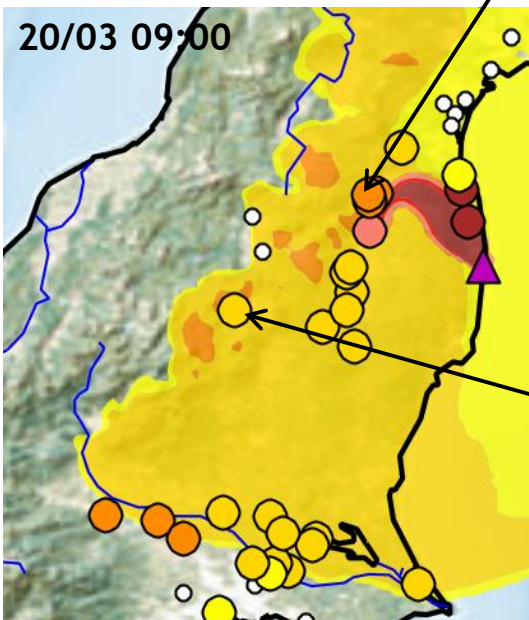
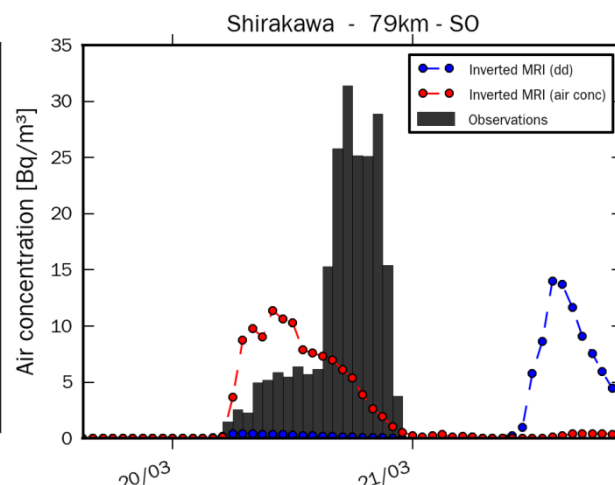
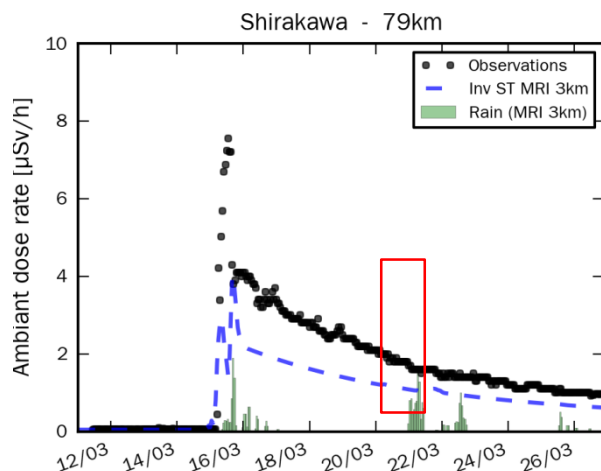
# Example of new perspectives bringing by the Tsuruta et al (2014) observations

## Plume 8 : 20/03 06:00 – 20/03 18:00

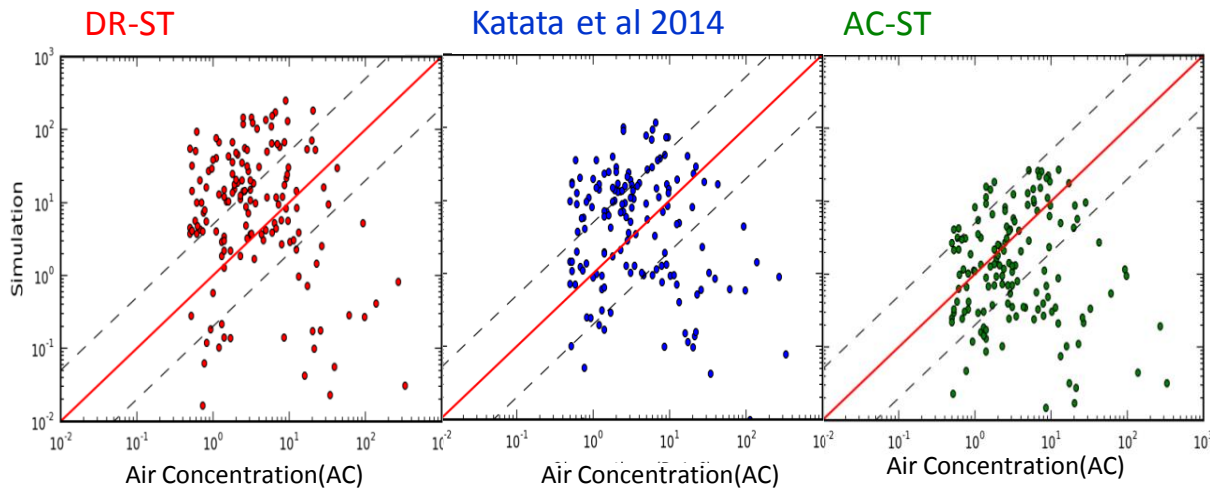
The morning of March 20, the plume direction was northward – high air concentrations was measured. In Miyagi prefecture (N), wet deposition was registered. Air concentrations measured in the surrounding areas were low (elevated plume?). Later that day, the plume moved gradually to the South of the Fuku. Pref.



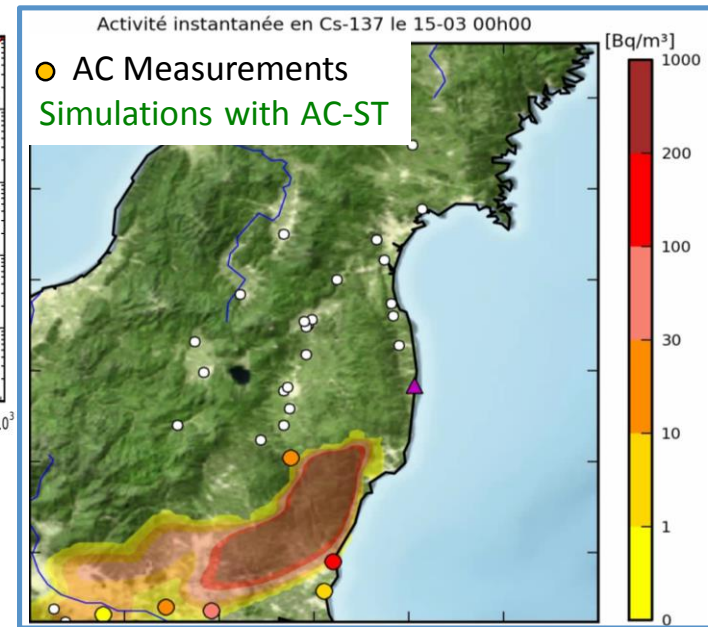
--- Simulations done with DR-ST --- Simulations done with AC-ST



Example : March 15 03:00 –18:00 UTC



	DR-ST	Katata et al 2014	AC-ST
FA5	30	40	54



- - - - Simulations done with DR-ST: (inverted ST assessed with Dose Rate meas. + MRI)
- - - - Simulations done with AC-ST : (inverted ST assessed with Tsuruta Air Concentration meas. + MRI)
- - - - Simulations done with Katata et al, 2015 ST

➤ Overestimation of the AC by simulating the event with the inverted DR-ST and Katata et al ST.

➤ Better agreement obtained for simulations done with AC-ST.

➤ Why is there such a difference?

# Inverse modelling approach to assess the source term

## □ Source-receptor relationship

$\mu$	=	$H$	=	$\sigma$	+	$\epsilon$
Vector of observations		Source receptor matrix computed with the forward ATM (Abida et al. 2011)		Estimator of the ST		Vector of errors Observations Model

## □ Variational method

$$J(\sigma) = \frac{1}{2}(\mu - H\sigma)^T R^{-1}(\mu - H\sigma) + \frac{1}{2}(\sigma - \sigma_b)^T B^{-1}(\sigma - \sigma_b)$$

- Cost function J which measures :
  - Differences between observations and model.
  - Differences between a priori and a posteriori emissions.
- Minimisation of J using gradient descent algorithm (L-BFGS-B method).

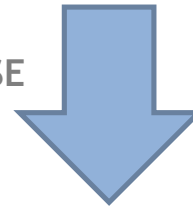
## □ Requirements

- Location of the accidental site.
- Number of observations >> number of unknowns parameters if *a priori*  $\sigma_b = 0$ .

## Features of the inversion process

ATM	IdX (C3X platform) Eulerian	No chemistry of iodine + Radioactive decay and filiation	Dry deposition $v_{\text{dep}} = 2 \cdot 10^{-3} \text{ cm/s}$	Wet deposition $\Lambda_s = a p_o$ $a = 5 \cdot 10^{-5}$	Vertical diffusion Troen and Mahrt scheme
Met. Data	MRI (IRSN/MRI collaboration)	Spatial resolution 0.03°	Time step 10 min	11/03/2011 - 01/04/2011 Rain: radar	
Observations	$^{137}\text{Cs}$ (Tsuruta) Dose rate	Temporal frequency : 1h		105 stations used for inversion 66 stations used for inversion	

INVERSE



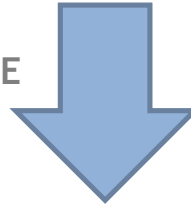
MODELING

Inverted ST	Temporal resolution	Radionuclides assessed	Period reconstruction
Tsuruta	1 hour	$^{137}\text{Cs}$	11/03 - 24/03
Dose rate	1 hour	$^{137}\text{Cs}$ , $^{134}\text{Cs}$ , $^{136}\text{Cs}$ , $^{137\text{m}}\text{Ba}$ , $^{131}\text{I}$ , $^{132}\text{I}$ $^{132}\text{Te}$ , $^{133}\text{Xe}$	11/03 - 27/03

## Features of the inversion process

ATM	IdX (C3X platform) Eulerian	No chemistry of iodine + Radioactive decay and filiation	Dry deposition $v_{\text{dep}} = 2 \cdot 10^{-3} \text{ cm/s}$	Wet deposition $\Lambda_s = ap_o$ $a = 5 \cdot 10^{-5}$	Vertical diffusion Troen and Mahrt scheme
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INVERSE MODELING



Inverted ST	Temporal resolution	Radionuclides assessed	Period reconstruction
Tsuruta	1 hour	$^{137}\text{Cs}$	11/03 - 24/03
Dose rate	1 hour	$^{137}\text{Cs}$ , $^{134}\text{Cs}$ , $^{136}\text{Cs}$ , $^{137\text{m}}\text{Ba}$ , $^{131}\text{I}$ , $^{132}\text{I}$ $^{132}\text{Te}$ , $^{133}\text{Xe}$	11/03 - 27/03

## Mission of IRSN in case of a Radiological Emergency

Assess risk induced by accidental situation - Provide technical expertise to public Authorities

Develop methods and tools to improve the operational response in case of an emergency

## The Fukushima accident : Understand, Reduce and Model the Uncertainties

The SAKURA project, an Hubert Curien Partnerships (PHC) between MRI and IRSN



### Related studies

- *Poster EGU2016-17692 Querel et al -> Wet deposition*
- *Poster EGU2016-4426 Perillat et al -> Modelling of uncertainties*



## Application with air concentration observations

JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 117, D05122, doi:10.1029/2011JD016932, 2012

### Estimation of errors in the inverse modeling of accidental release of atmospheric pollutant: Application to the reconstruction of the cesium-137 and iodine-131 source terms from the Fukushima Daiichi power plant

Victor Winiarek,<sup>1,2</sup> Marc Bocquet,<sup>1,2</sup> Olivier Saunier,<sup>3</sup> and Anne Mathieu<sup>3</sup>

Received 27 September 2011; revised 19 January 2012; accepted 23 January 2012; published 9 March 2012.

## Application with dose rate observations

Atmos. Chem. Phys., 13, 11403–11421, 2013

www.atmos-chem-phys.net/13/11403/2013/

doi:10.5194/acp-13-11403-2013

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Atmospheric  
Chemistry  
and Physics



### An inverse modeling method to assess the source term of the Fukushima Nuclear Power Plant accident using gamma dose rate observations

O. Saunier<sup>1,\*</sup>, A. Mathieu<sup>1</sup>, D. Didier<sup>1</sup>, M. Tombette<sup>1</sup>, D. Quélo<sup>1</sup>, V. Winiarek<sup>2,3</sup>, and M. Bocquet<sup>2,3</sup>

## Application with air concentration and deposition observations

Atmospheric Environment 82 (2014) 268–279



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journal homepage: [www.elsevier.com/locate/atmosenv](http://www.elsevier.com/locate/atmosenv)



Estimation of the caesium-137 source term from the Fukushima Daiichi nuclear power plant using a consistent joint assimilation of air concentration and deposition observations

Victor Winiarek<sup>a,b,\*</sup>, Marc Bocquet<sup>a,b</sup>, Nora Duhanyan<sup>a</sup>, Yelva Rouston<sup>a</sup>, Olivier Saunier<sup>c</sup>, Anne Mathieu<sup>c</sup>



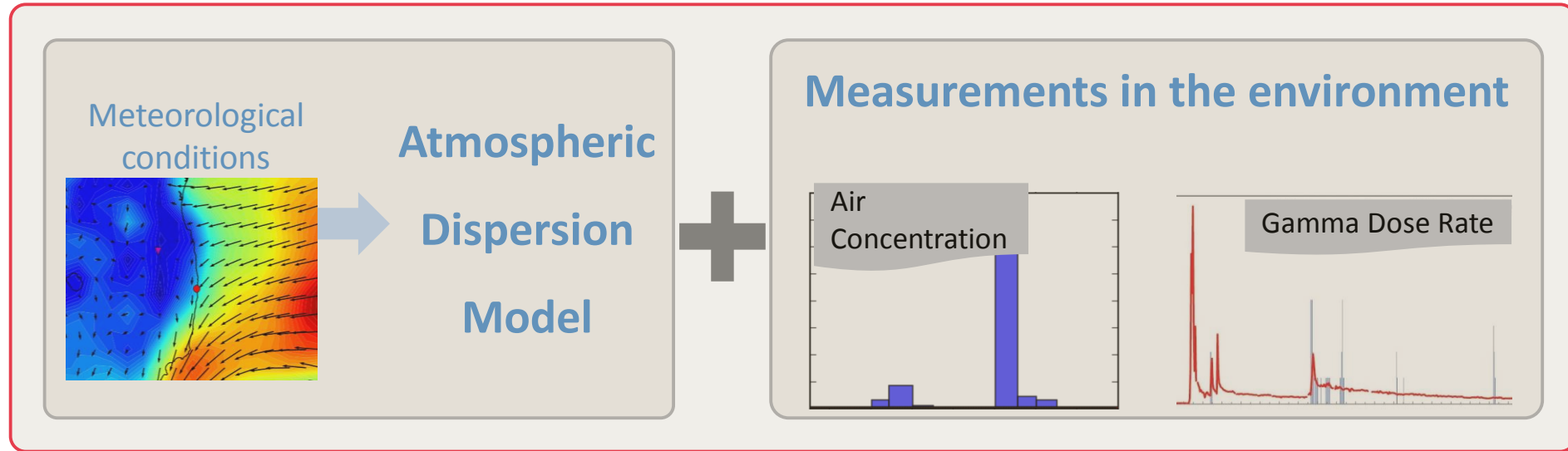
**Operational**

Still some improvements need to be done

**Not Operational**

First attempt to take into account several kind of data: promising results but not yet suited for operational use.

# Tool to assess the Source Term by using observations in the environment

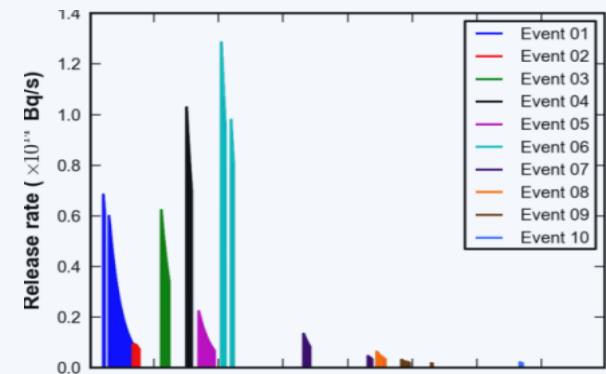


**Inverse modelling approach**

(mathematical rigorous approach)

## Source term estimate

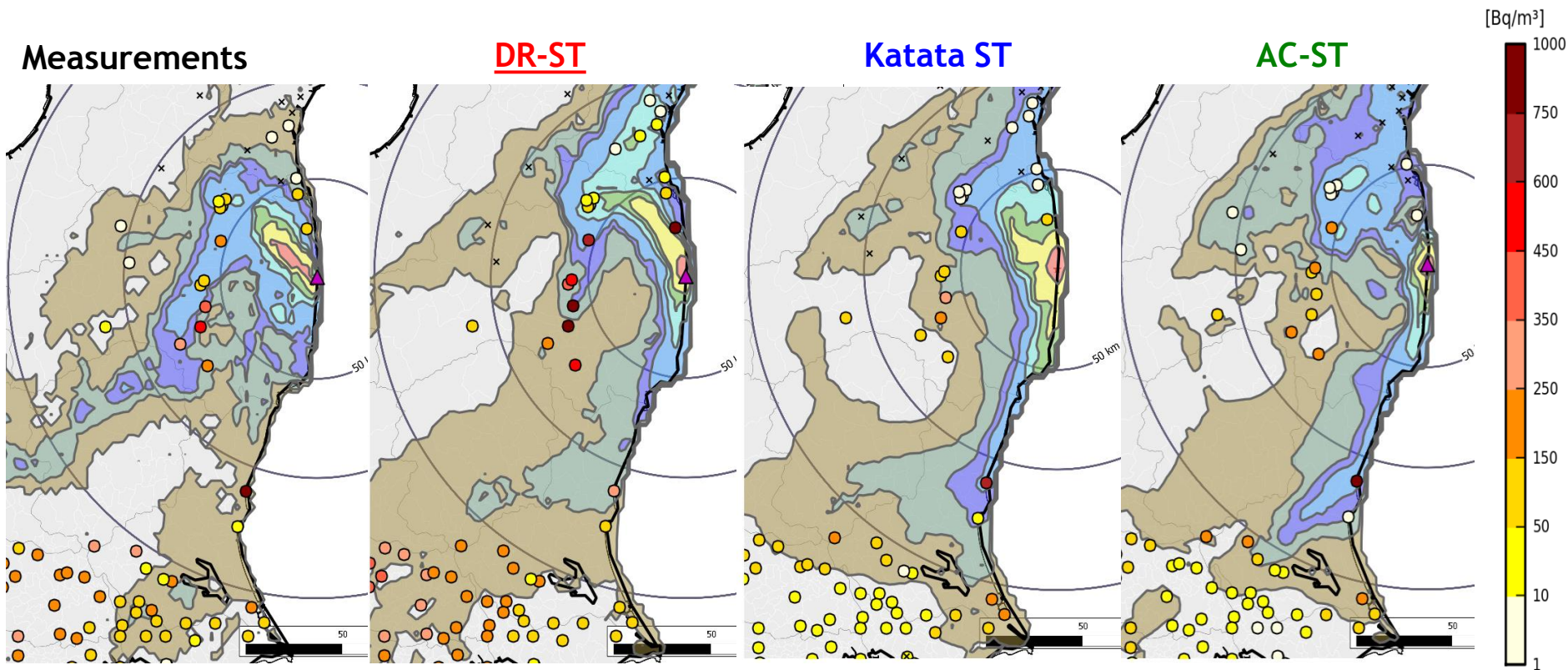
The compromise providing the best agreement between model and observations



*ST = temporal evolution of the release rate + distribution between radionuclides*

## Example : March 15 03:00 –18:00 UTC

Total  $^{137}\text{Cs}$  +  $^{134}\text{Cs}$  deposition and cumulated  $^{137}\text{Cs}$  air concentration during the event



- Only DR-ST simulation partially reconstitutes the deposition pattern in the Nakadori valley.
- But DR-ST simulation significantly over-estimates the cumulated AC in the valley.

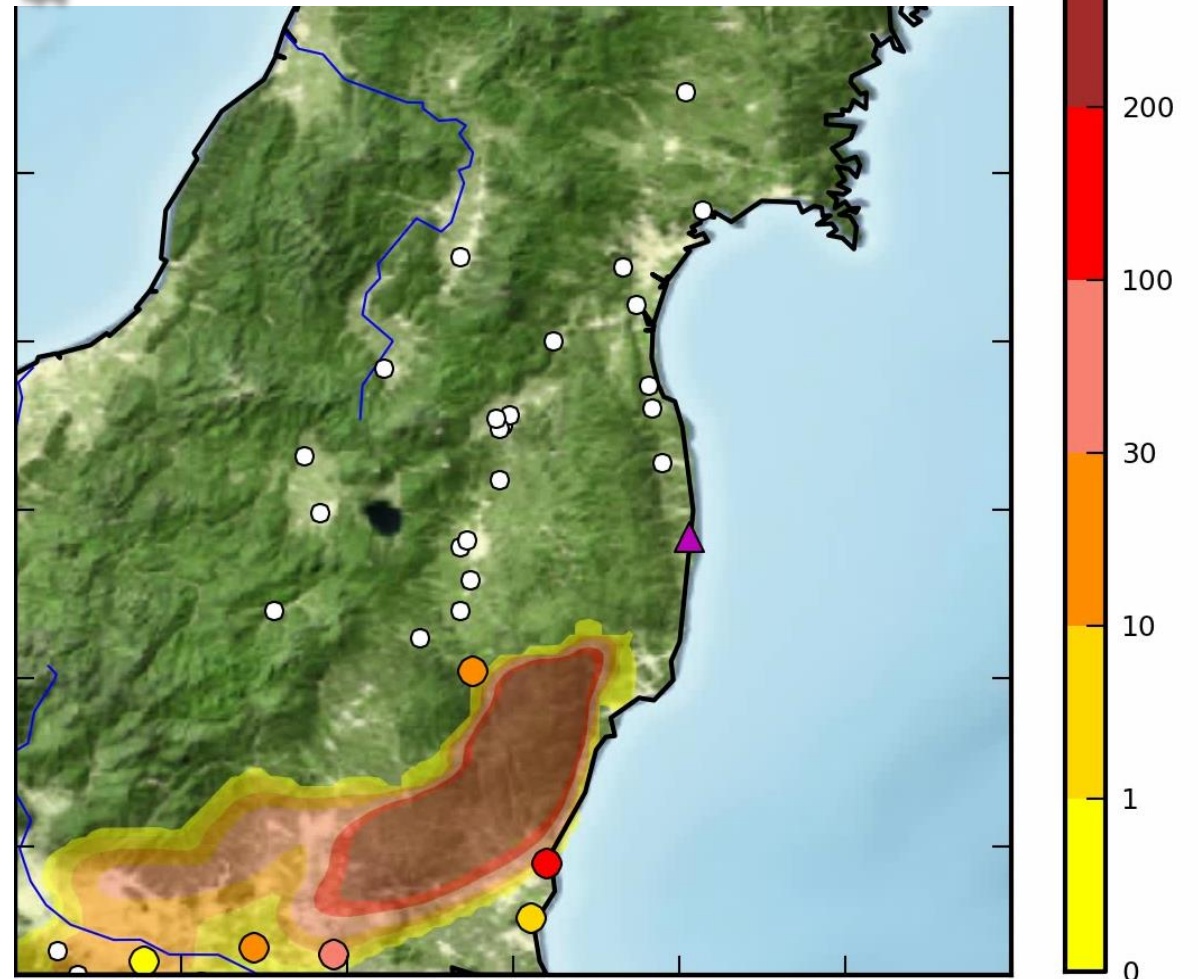
## Deposition in the Nakadori valley and in the NW of the FDNPP occurred on March 15 01:00 –18:00 UTC

- Air concentration measurements
- **B** Plume simulation with inverted air concentration ST [Bq/m<sup>3</sup>]
- Radar rain observations

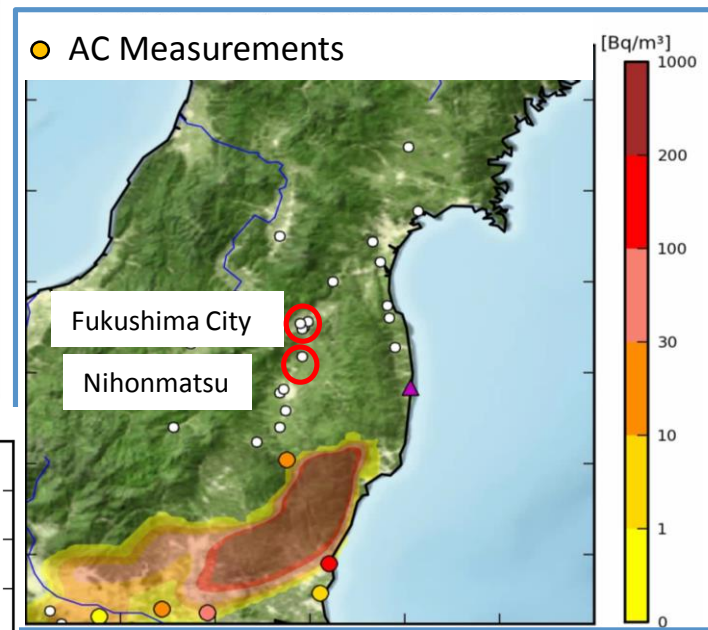
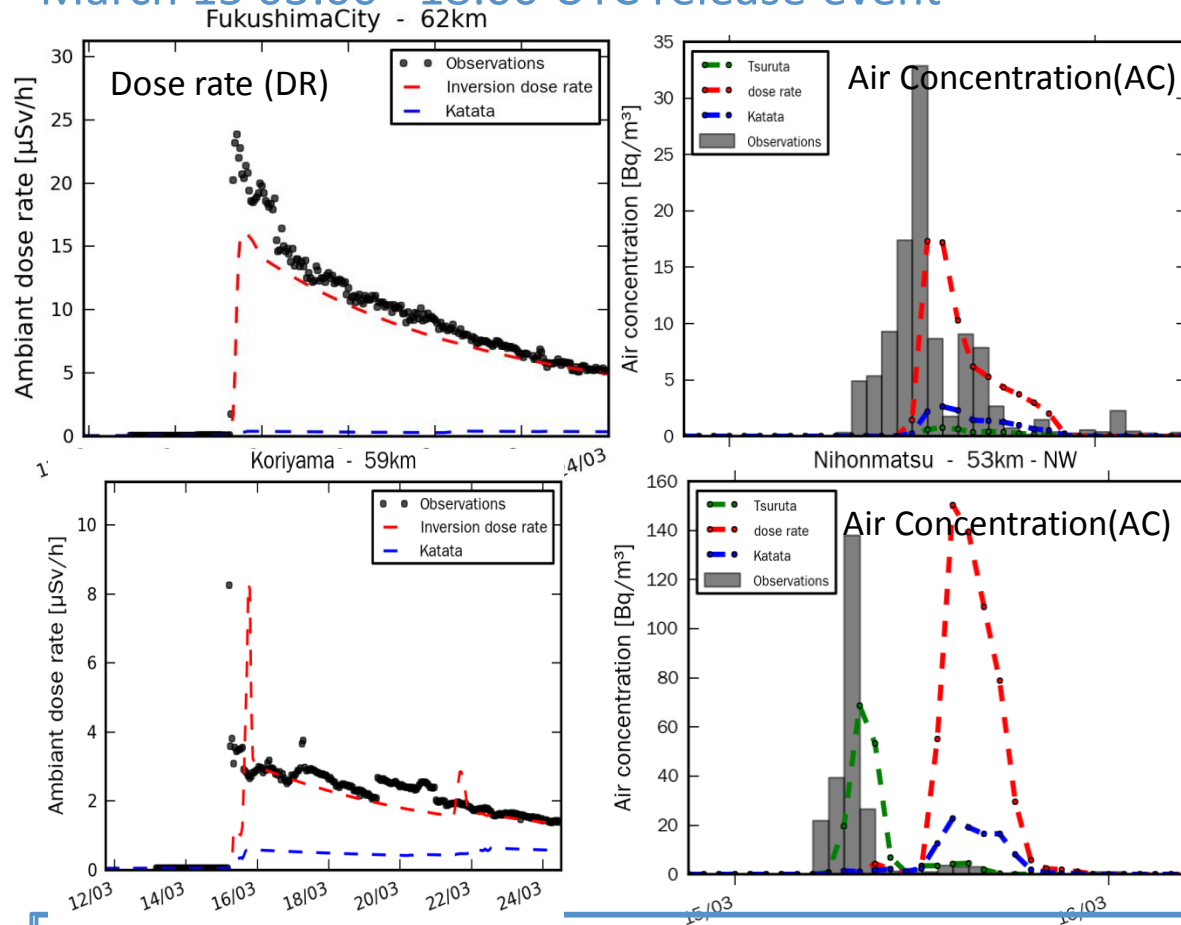
The wind gradually turned advecting releases first southward (0h) then south-westward (2h), westward (5h), north-westward (8h).

Wet deposition started at ~7h in the Nakadori valley and at ~8h in the north-west area.

At noon, wind gradually turned back. Plumes were advected northwestward then westward and southward.



# March 15 03:00 –18:00 UTC release event

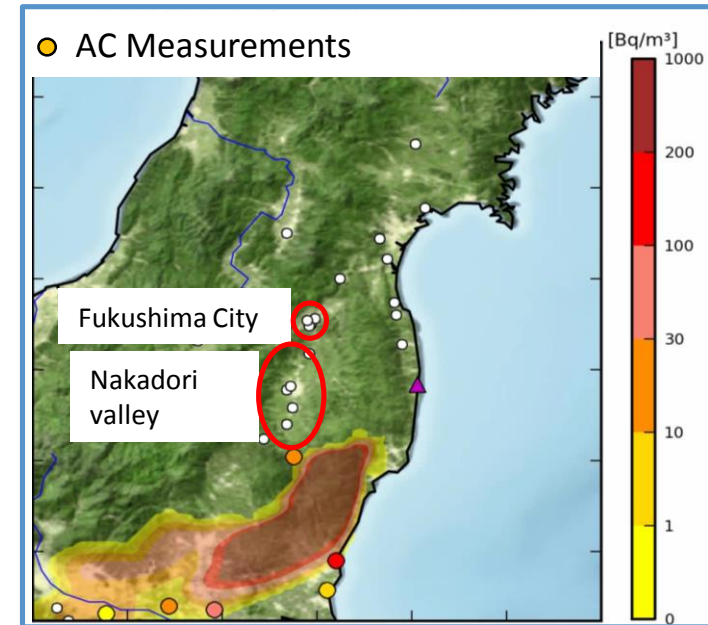
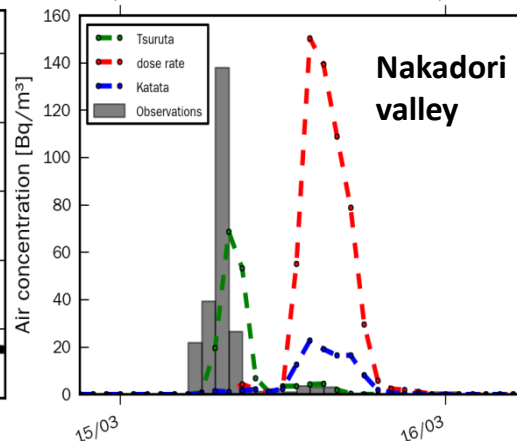
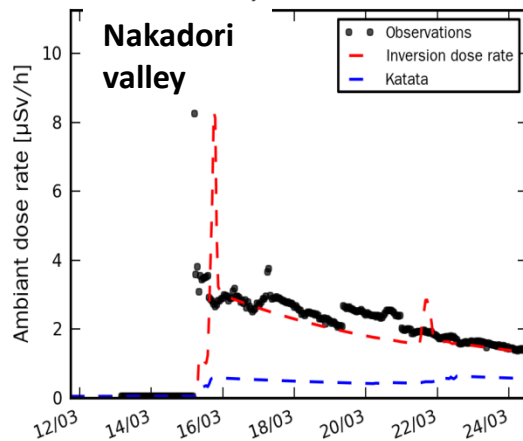
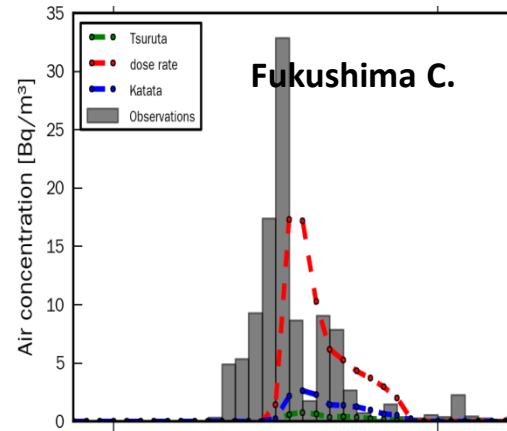
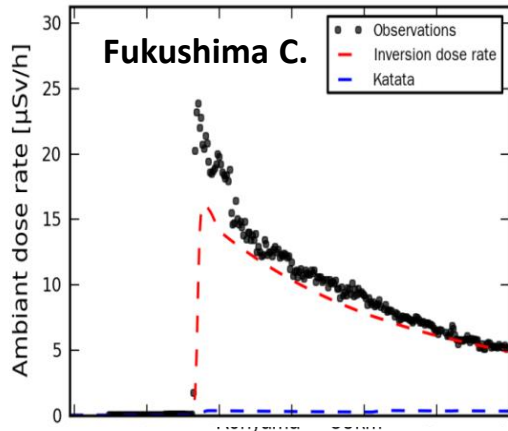


- - - **A** Simulation done with the inverted ST assessed with Dose Rate.
- - - **B** Simulation done with the inverted ST assessed with Tsuruta Air Concentration.
- - - Simulation done with Katata et al, 2015 ST

## Fukushima city:

- A** simulation of the **A** rcity with the observed DR and the observed AC but delayed (because of what happened in Fukushima city). Same tendency with Katata simulation
- B** and Katata simulation does not correctly simulate
- B** agrees correctly with observed AC

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--- **A** Simulation done with the inverted ST assessed with Dose Rate.

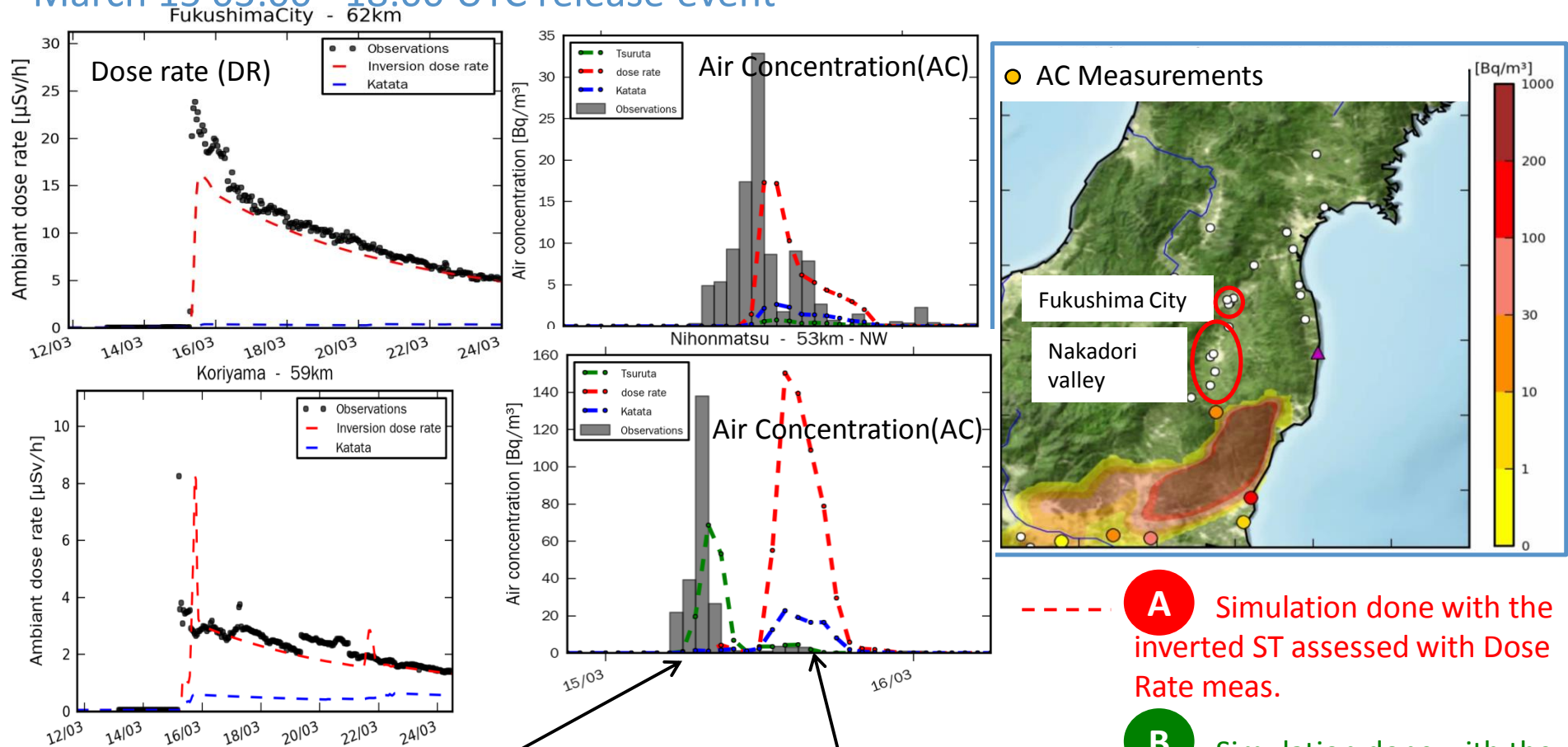
--- **B** Simulation done with the inverted ST assessed with Tsuruta Air Concentration.

--- Simulation done with Katata et al, 2015 ST

According to the observations the deposit observed in the Nakadori valley occurred mainly during this event: wet deposition probably occurred during a light rain event

the deposit in the Nakadori valley is simulated with the inverted dose rate ST during this event (precipitations are observed and simulated).

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the deposit in the Nakadori valley is simulated with the inverted dose rate ST during this event (precipitations are observed and simulated).

**A** Simulation done with the inverted ST assessed with Dose Rate meas.

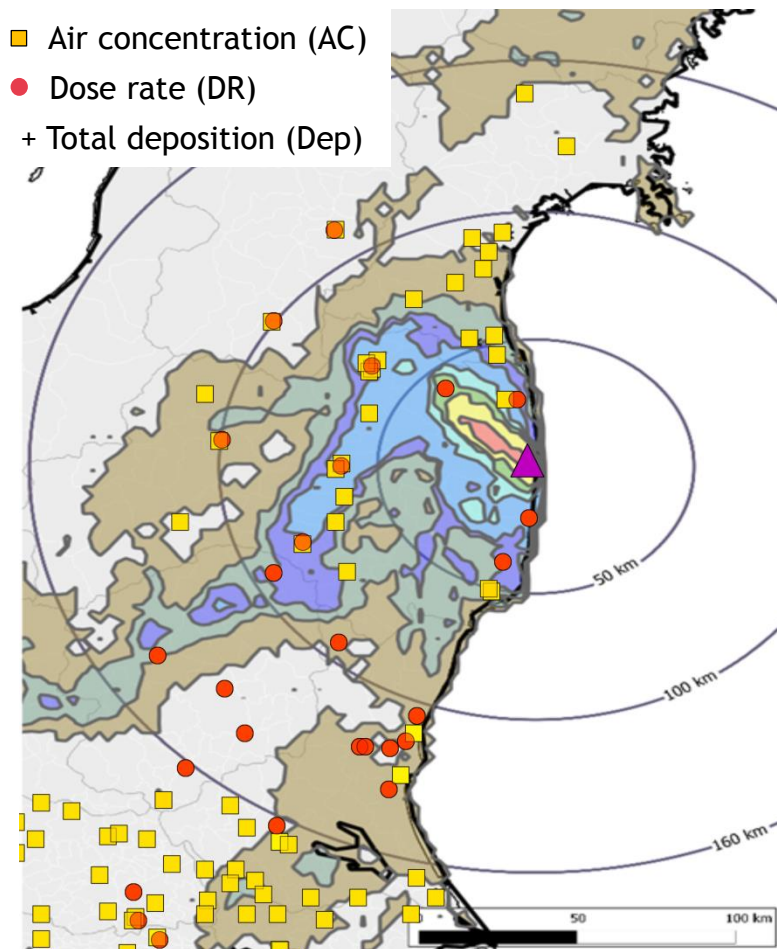
**B** Simulation done with the inverted ST assessed with Tsuruta Air Concentration meas.

Simulation done with Katata et al, 2015 ST

# Model to data comparison with the Tsuruta et al (2014) observations

Air Pollution Surveillance Network provided  $^{137}\text{Cs}$  air concentration measurements with an hourly temporal time step (11520 obs.).

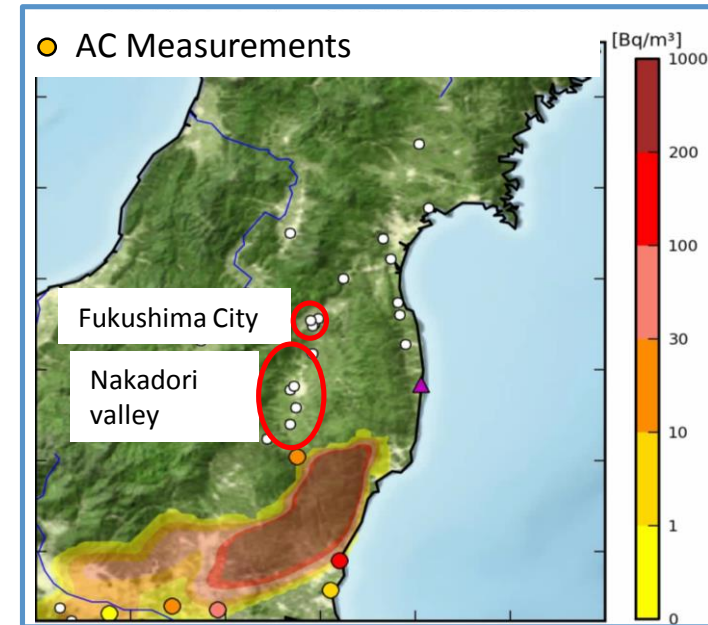
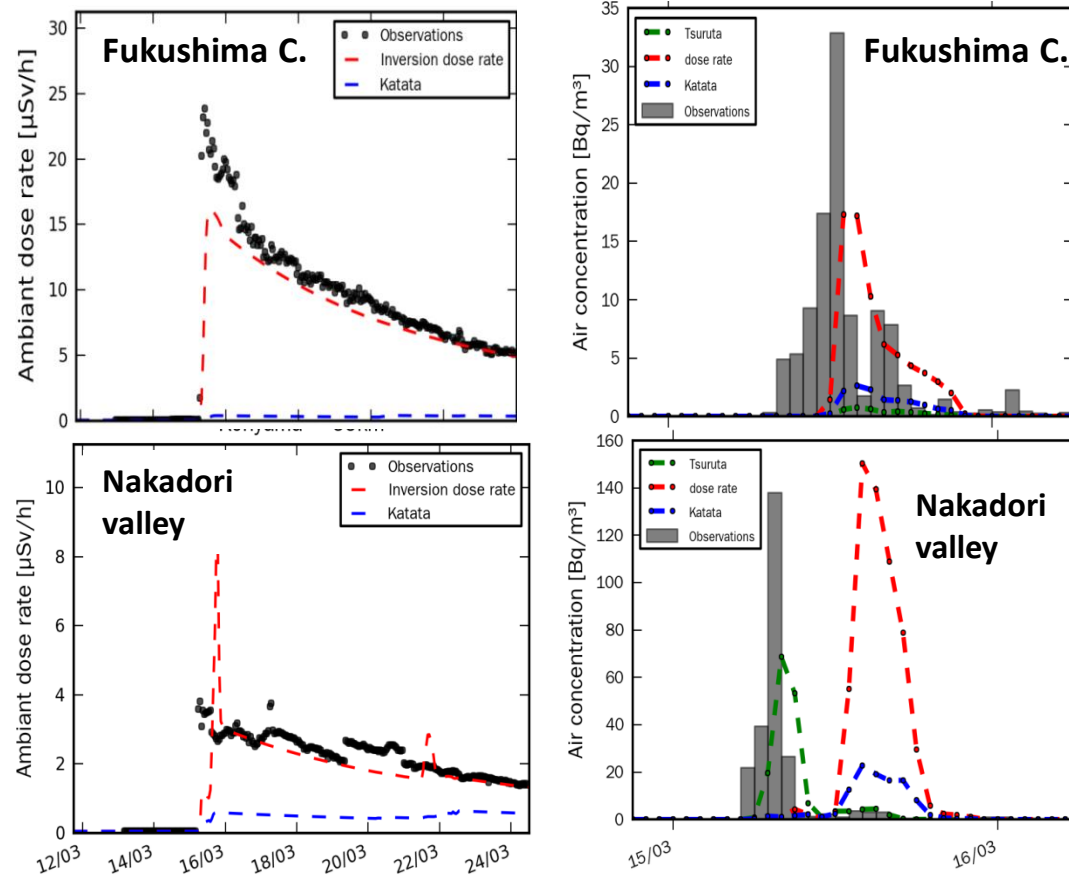
Data available during 4 periods of time: March 12-13, 15-16, 18-19, 20-23



- ❑ No station in the main contaminated area (NW).
- ❑ Several stations N of FDNPP (Dry deposition on 03/12).
- ❑ Several stations in the center of Fukushima prefecture. (Wet dep. on 03/15 - fog??).
- ❑ Several stations in the border between Ibaraki prefecture and Chiba prefecture. (wet dep. du 21/03).



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- - - Simulation done with Katata et al, 2015 ST

### ➔ Fukushima city:

- **A** simulation agrees correctly with the observed DR and the observed AC but simulated plume is slightly delayed
- **B** and Katata simulations do not correctly simulate the plume