CI Congresso Nazionale della Società Italiana di Fisica

September 21st 2015, Rome - ITALY

# Radioxenon Atmospheric Transport Modeling: From Worldwide Impact of Nuclear Power Plants to CTBTO Event Screening Categorization

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# Motivitation

- Comprehensive Nuclear Test Ban Treaty Organization (CTBTO)

  *International Noble Gas Experiment - virtual Data Exploitation Centre Collaboration*

- Atmospheric Transport Modelling (ATM)

  *European Centre for Medium-Range Weather Forecasts*

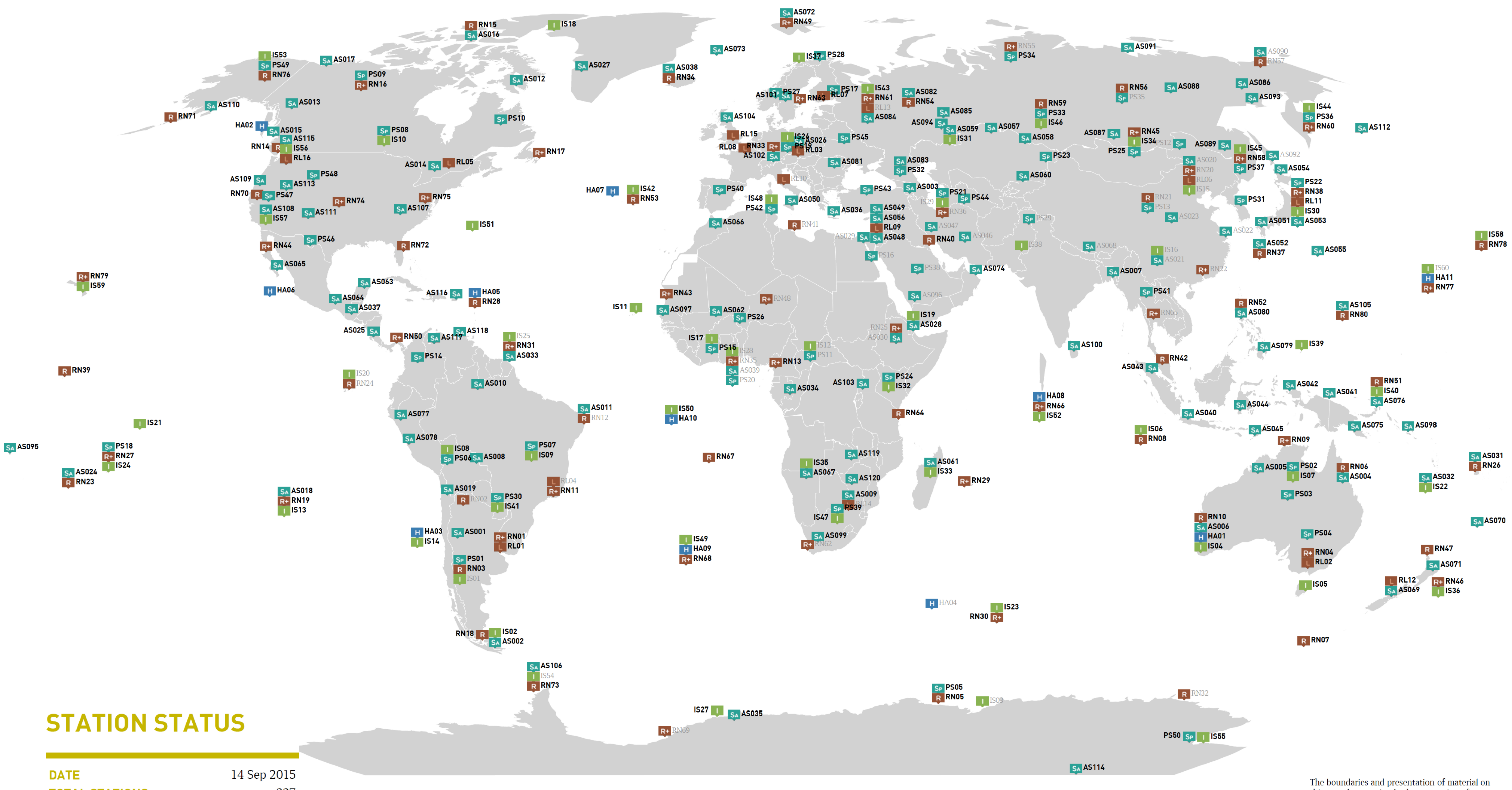
- Meteorological Pattern Analysis and Radioxenon Emission Inventory

# INTERNATIONAL MONITORING SYSTEM

## GLOBAL OVERVIEW - CERTIFIED STATIONS AND NON-CERTIFIED STATIONS

**14 SEPTEMBER 2015**

CTBTO PREPARATORY COMMISSION — preparatory commission for the comprehensive nuclear-test-ban treaty organization



### STATION STATUS

| | |
|---|---|
| DATE | 14 Sep 2015 |
| TOTAL STATIONS | 337 |
| PLANNING | 18 |
| UNDER CONSTRUCTION | 18 |
| INSTALLED | 20 |
| CERTIFIED | 281 |

SP Primary Seismic · SA Auxiliary Seismic · I Infrasound · H Hydroacoustic · R Radionuclide · R+ Radionuclide w/ Noble Gas · L Radionuclide Lab

The boundaries and presentation of material on this map does not imply the expression of any opinion on the part of the Provisional Technical Secretariat concerning the legal status of any country, territory, city or area or its authorities, or concerning the delimitation of its frontiers or boundaries.

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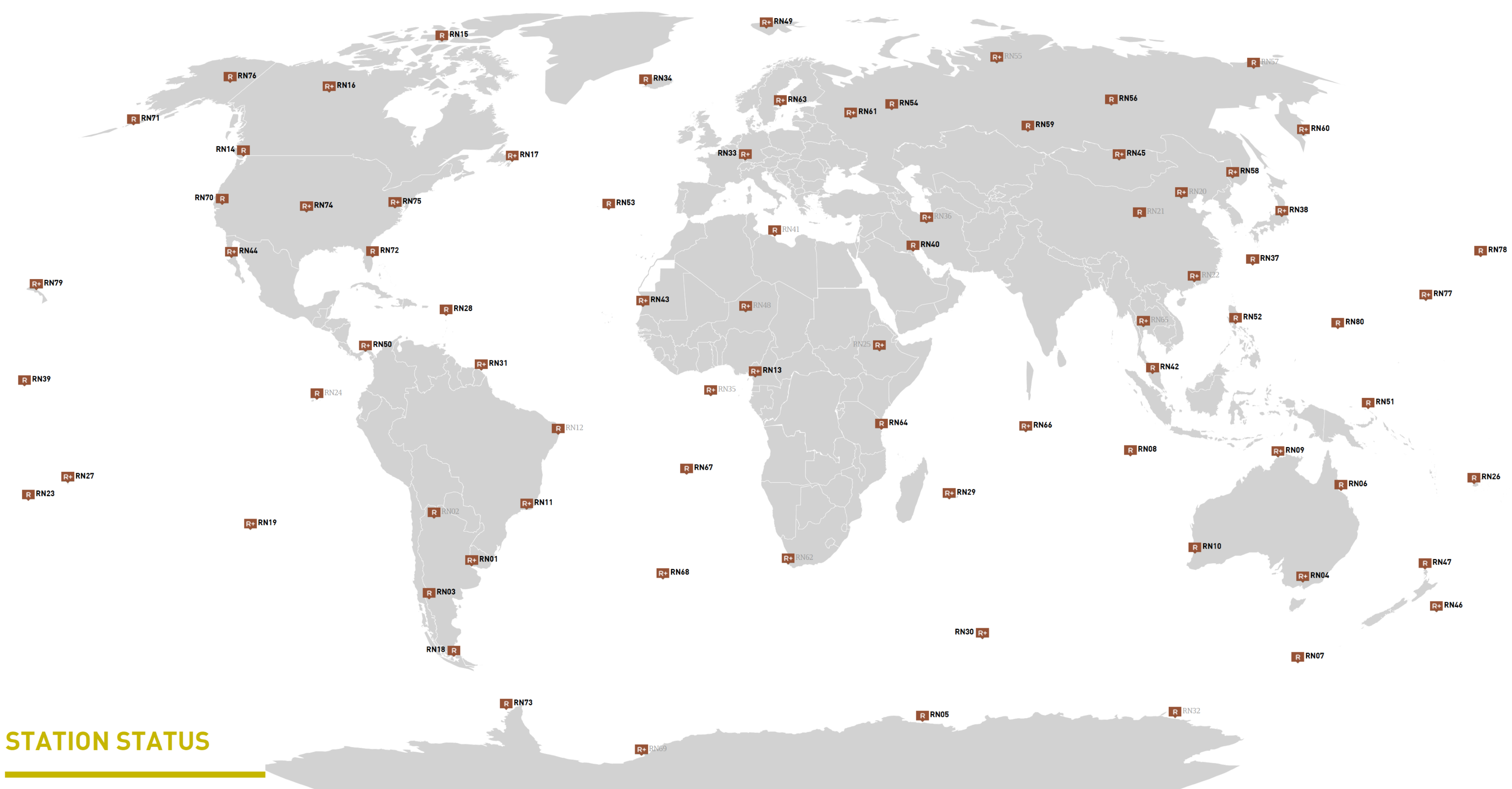
## INTERNATIONAL MONITORING SYSTEM

### RADIONUCLIDE NETWORK - CERTIFIED STATIONS AND NON-CERTIFIED STATIONS

14 SEPTEMBER 2015

CTBTO PREPARATORY COMMISSION | preparatory commission for the comprehensive nuclear-test-ban treaty organization




## STATION STATUS

| | |
|---|---|
| DATE | 14 Sep 2015 |
| TOTAL STATIONS | 80 |
| PLANNING | 4 |
| UNDER CONSTRUCTION | 10 |
| INSTALLED | 3 |
| CERTIFIED | 63 |

SP Primary Seismic · SA Auxiliary Seismic · I Infrasound · H Hydroacoustic · R Radionuclide · R+ Radionuclide w/ Noble Gas · L Radionuclide Lab

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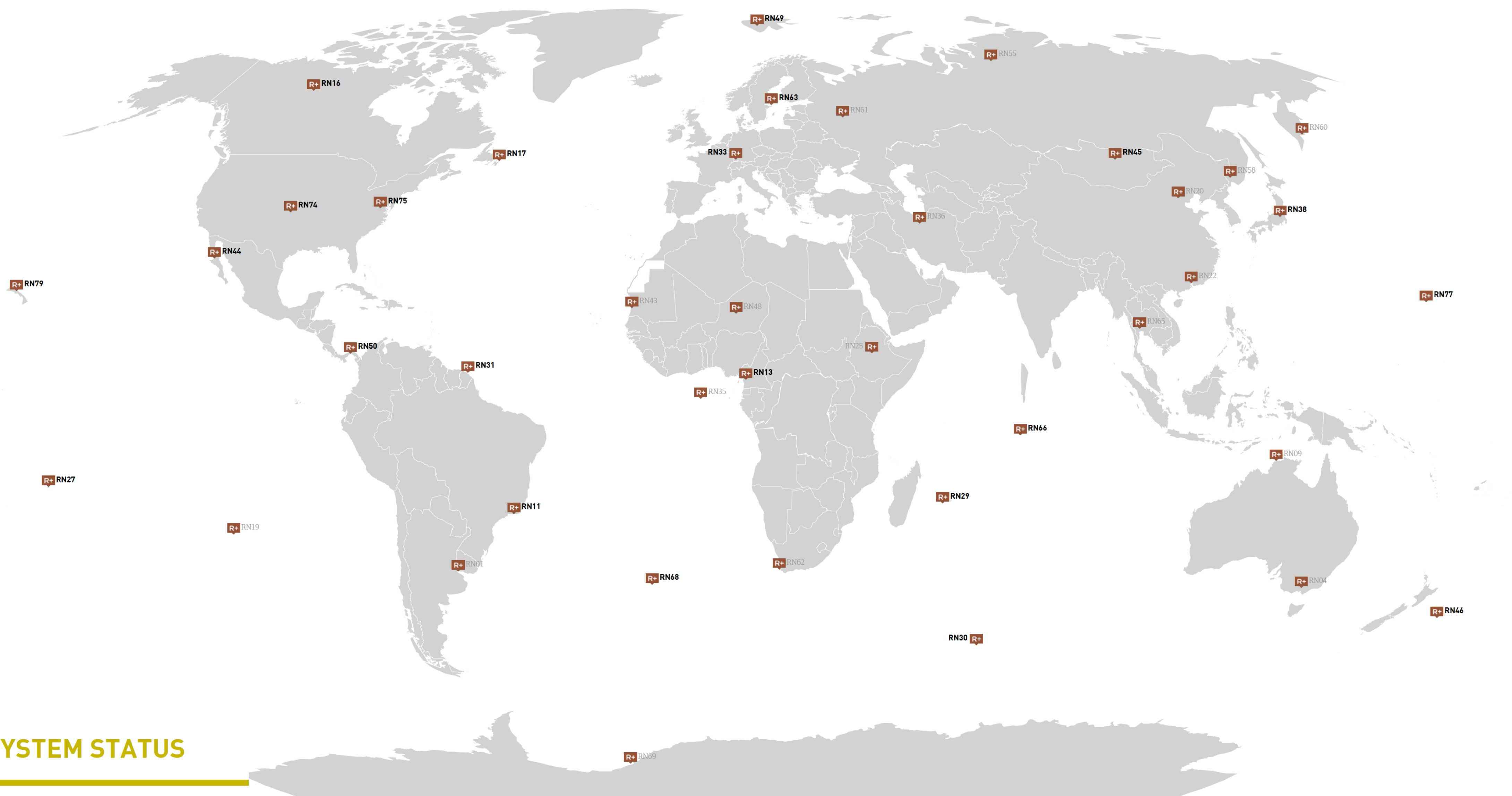
## INTERNATIONAL MONITORING SYSTEM

### NOBLE GAS NETWORK - CERTIFIED SYSTEMS AND NON-CERTIFIED SYSTEMS

**14 SEPTEMBER 2015**

CTBTO PREPARATORY COMMISSION | preparatory commission for the comprehensive nuclear-test-ban treaty organization




## SYSTEM STATUS

| | |
|---|---|
| DATE | 14 Sep 2015 |
| TOTAL SYSTEMS | 40 |
| PLANNING | 0 |
| UNDER CONSTRUCTION | 10 |
| INSTALLED | 8 |
| CERTIFIED | 22 |

SP Primary Seismic · SA Auxiliary Seismic · I Infrasound · H Hydroacoustic · R Radionuclide · R+ Radionuclide w/ Noble Gas · L Radionuclide Lab

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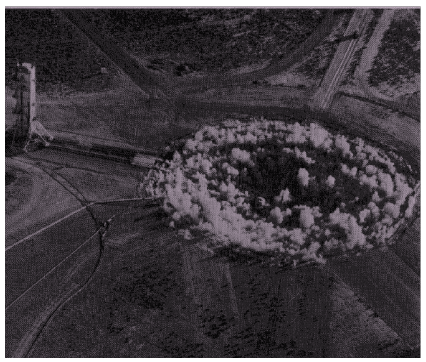


Atmospheric transport to station
1-14 days or longer

IMS station

| Particulate | Noble gas |
| --- | --- |
| Sample collection | |
| 24 hours | 12-24 hours |
| Decay/sample processing | |
| 24 hours | 0-7 hours |
| Data acquisition | |
| 24 hours | 12-24 hours |
| Data is ready | |
| 3 days | ~2 days |

Delay: 2 – 14 days or longer

IDC, Vienna

Transmissions by GCI

NDC's

**Argon-37:** activation product. Decays with e.c.

**Krypton-85** ($t_{1/2}$= 10.7 y): world-wide high background

**Xenon's**: four isotopes are suitable for CTBT verification ($^{131m}Xe$, $^{133m}Xe$, $^{133}Xe$, and $^{135}Xe$)

**Argon-37:** activation product. Decays with e.c.

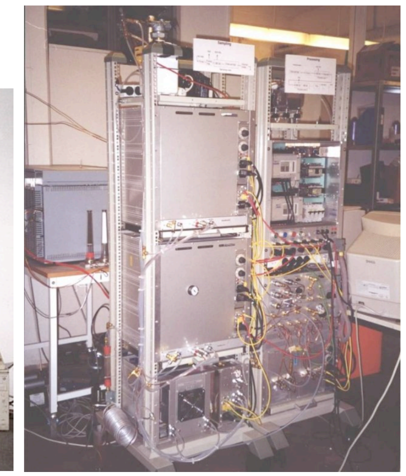
**Krypton-85** ($t_{1/2}$= 10.7 y): world-wide high background

**Xenon's**: four isotopes are suitable for CTBT verification ($^{131m}Xe$, $^{133m}Xe$, $^{133}Xe$, and $^{135}Xe$)

| Fission Product | Half-life | Time unit | $^{235}U_f$ | $^{235}U_{he}$ | $^{238}U_f$ | $^{238}U_{he}$ | $^{239}Pu_f$ | $^{239}Pu_{he}$ |
|---|---|---|---|---|---|---|---|---|
| $^{131m}Xe$ | 11.934 | d | 0.05 | 0.06 | 0.05 | 0.06 | 0.05 | 0.07 |
| $^{133m}Xe$ | 2.19 | d | 0.19 | 0.29 | 0.19 | 0.18 | 0.24 | 0.42 |
| $^{133}Xe$ | 5.243 | d | 6.72 | 5.53 | 6.76 | 6.02 | 6.97 | 4.86 |
| $^{135}Xe$ | 9.14 | h | 6.6 | 5.67 | 6.97 | 5.84 | 7.54 | 6.18 |

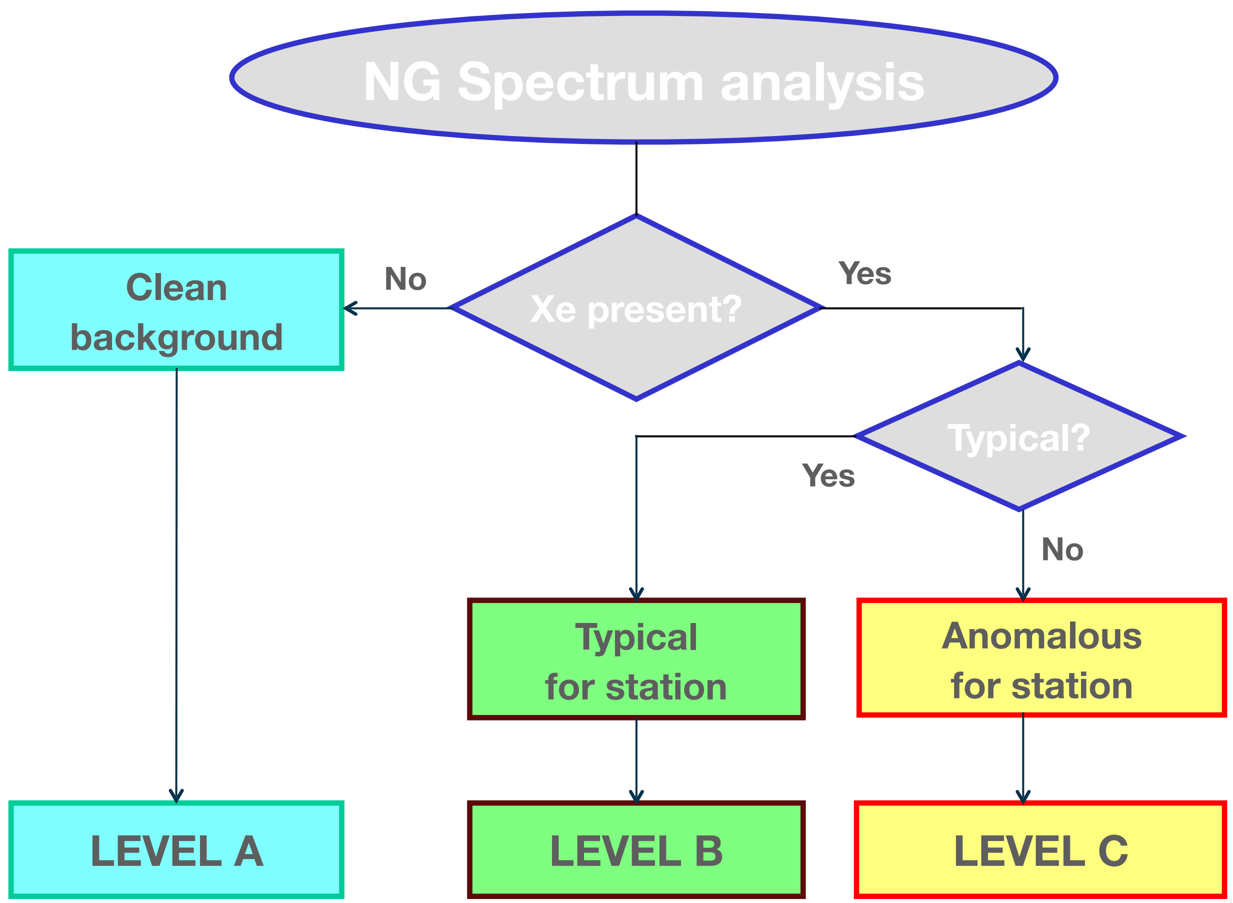
- Cumulative fission yields in % for six fission modes relevant to nuclear explosions, induced by fission spectrum neutrons (f) and high energy neutrons (14.7 MeV) (he)

- $^{133}Xe$ has high production rates and a not too short half-life. Therefore this xenon isotope is the one most observed in environmental samples




| Characteristics | Minimum requirements | SPALAX | SAUNA | ARIX |
|---|---|---|---|---|
| Airflow | 0.4 $m^3/h$ | 15–20 $m^3/h$ | 1.2 $m^3/h$ | 1.6 $m^3/h$ |
| Total volume of sample | 10 $m^3$ | 50–75 $m^3$ | 25–30 $m^3$ | 36 $m^3$ |
| Collection time | ≤24 h | ≤24 h | 12 h | 12 h |
| Measurement time | ≤24 h | ≤24 h | 11 h 10 min | 18 h |
| Time before reporting | ≤48 h | ≤48 h | 30 h | 34 h |
| Reporting frequency | Daily | Daily | 2 samples per day | 2 samples per day |
| Isotopes measured | $^{131m}$Xe, $^{133}$Xe, $^{133m}$Xe, $^{135}$Xe | $^{131m}$Xe, $^{133}$Xe, $^{133m}$Xe, $^{135}$Xe | $^{131m}$Xe, $^{133}$Xe, $^{133m}$Xe, $^{135}$Xe | $^{131m}$Xe, $^{133}$Xe, $^{133m}$Xe, $^{135}$Xe |
| Measurement mode | $\beta$-$\gamma$-coincidence or high-res-$\gamma$- spectrometry | High-res-$\gamma$- spectrometry | $\beta$-$\gamma$-coincidence | $\beta$-$\gamma$-coincidence |
| MDC for $^{133}$Xe | 1 $mBq/m^3$ | 0.2–0.6 $mBq/m^3$ | 0.2–0.4 $mBq/m^3$ | 0.2–0.3 $mBq/m^3$ |

Auer, M. *et al*. (2010). Ten years of development of equipment for measurement of atmospheric radioactive xenon for the verification of the CTBT. Pure and Applied Geophysics 167, 471-486.




**Categorization Levels:**

Concentrations
Xe-133m
Xe-131m
Xe-135
Xe-133

**ATM Flag:**

Backtracking indicates known source

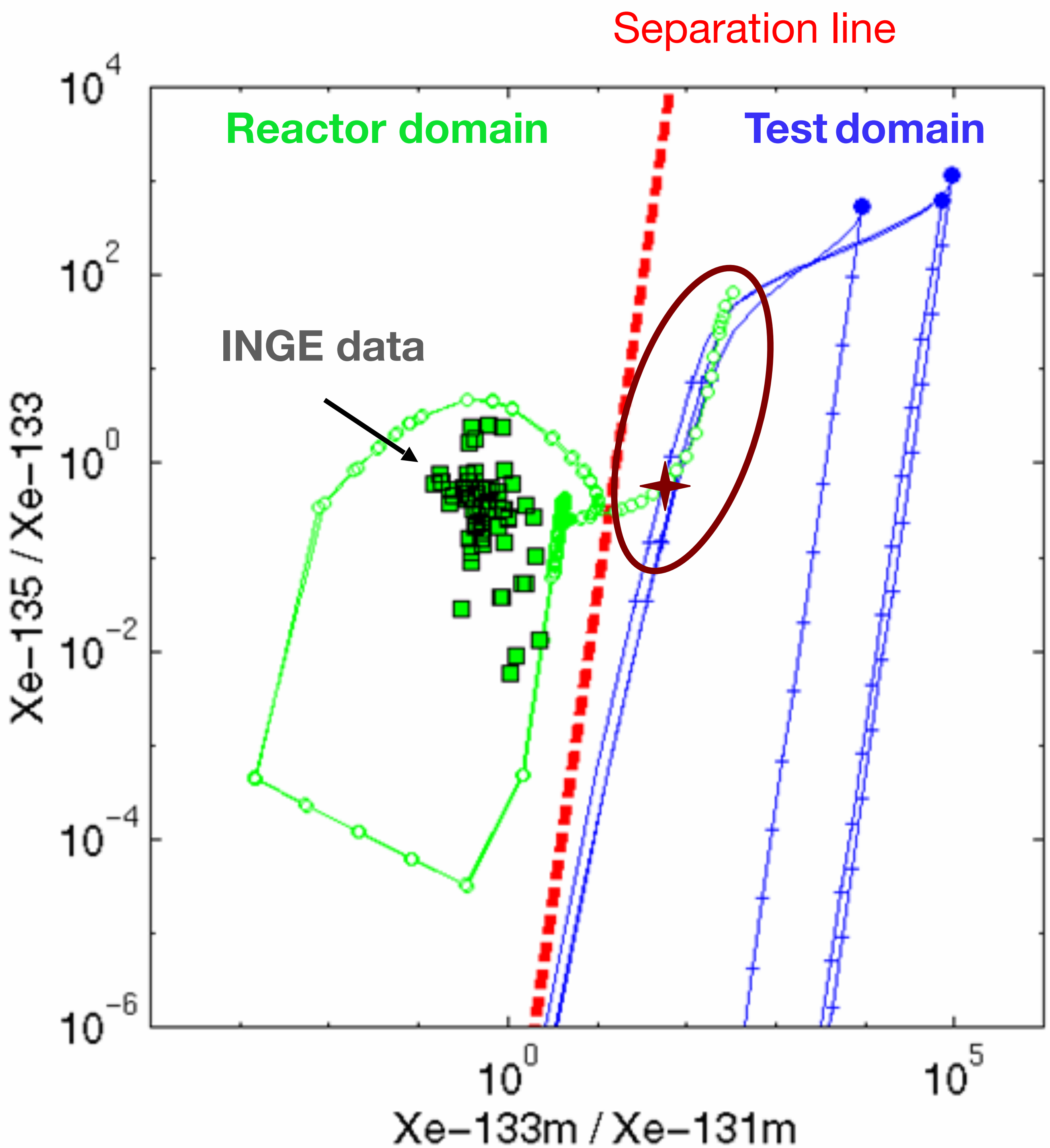
**Short term Flag:**

Short term Category

**SOH Flag:**

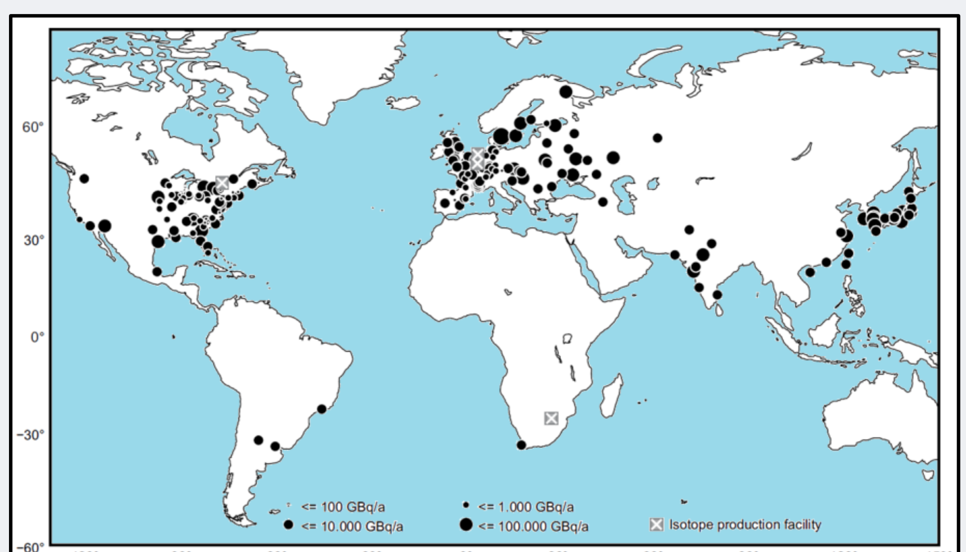
Quality Indicators On Sample reliability

**Flag:**
Isotopic ratios
Xe-133m/131m > 2?
Xe-135/133 > 5?
Xe-133m/Xe-133 > 0.3?

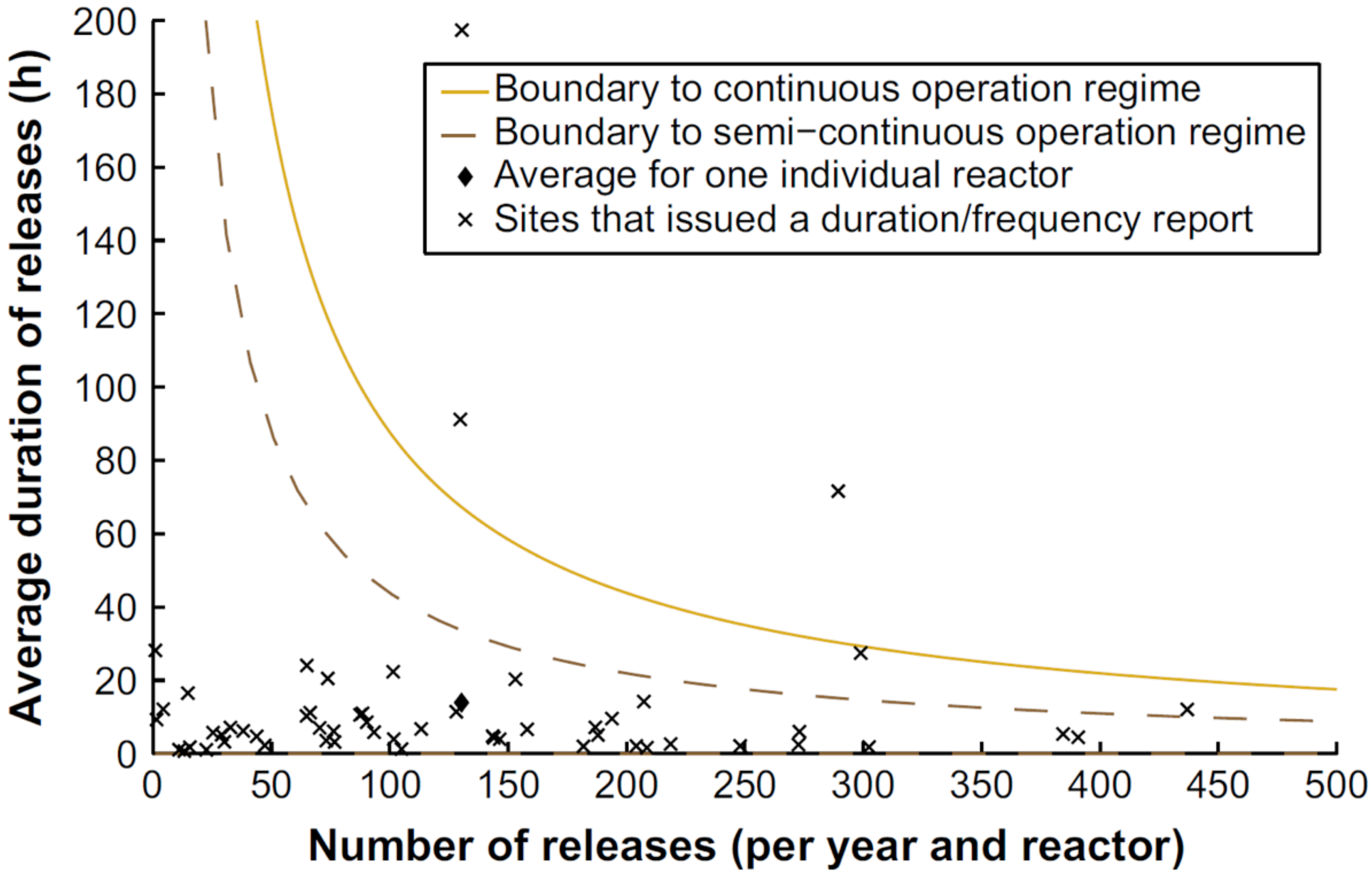

Separation line
Reactor domain
Test domain
INGE data
$10^4$
$10^2$
$10^0$
$10^{-2}$
$10^{-4}$
$10^{-6}$
Xe−135 / Xe−133
$10^0$
$10^5$
Xe−133m / Xe−131m

| Nuclear explosions | Civil background sources of $^{133}Xe$ | | |
|---|---|---|---|
| | Isotope production facilities | Nuclear power plants | Other sources |
| • Induced fission of $^{235}U$ or $^{239}Pu$<br>• Reaction time <1μs<br>• About **$10^{16}$ Bq** of $^{133}Xe$ in a 1kt TNT equivalent explosion<br>• In underground scenario leakage of ≈1-10% | • 5 facilities<br>• Supply of $^{99}Mo$<br>• **$10^{12}$ Bq/day** | • ≈ 200 facilities<br>• Energy supply<br>• **$10^{10}$ Bq/day** | Nuclear reactions<br>• Research reactors<br>• Nuclear ships and submarines<br>• Nuclear waste reprocessing<br>• Hospitals |

Heterogenic distribution in the world:




Too small and irregular




Number and duration of NPP radioxenon batch releases. Each mark represents one NPP site with available emission reports from November 2007. Only few operate in the continuous operation regime.

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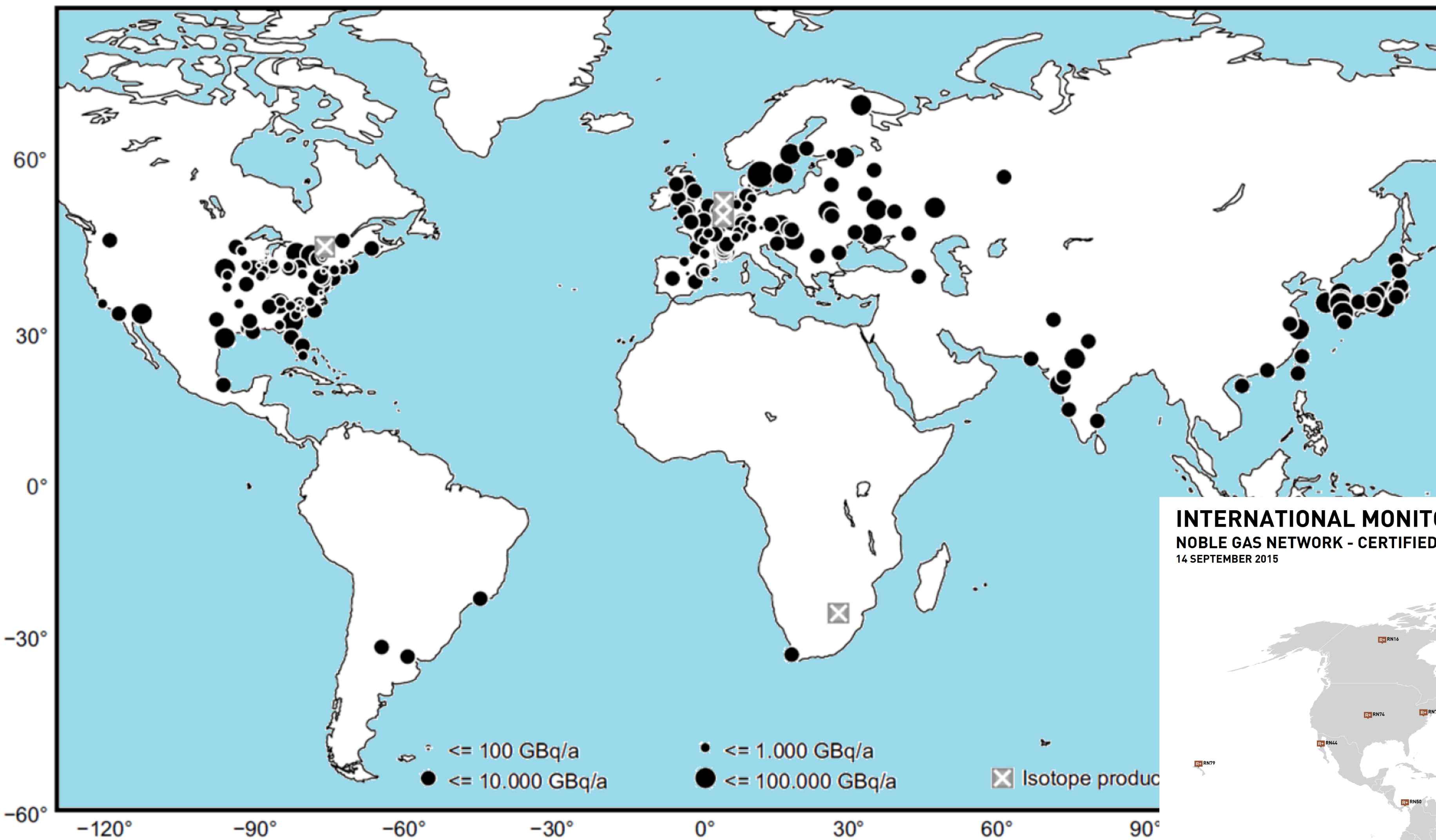
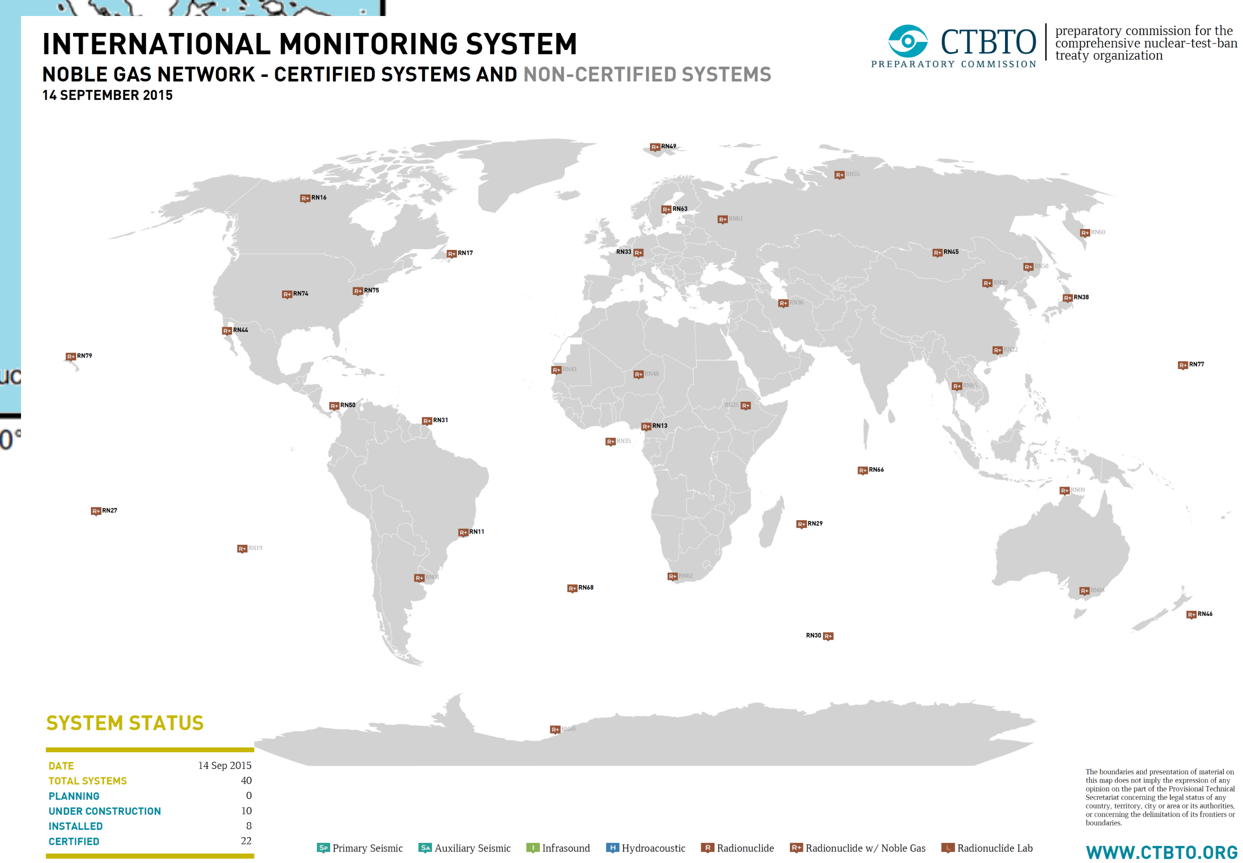


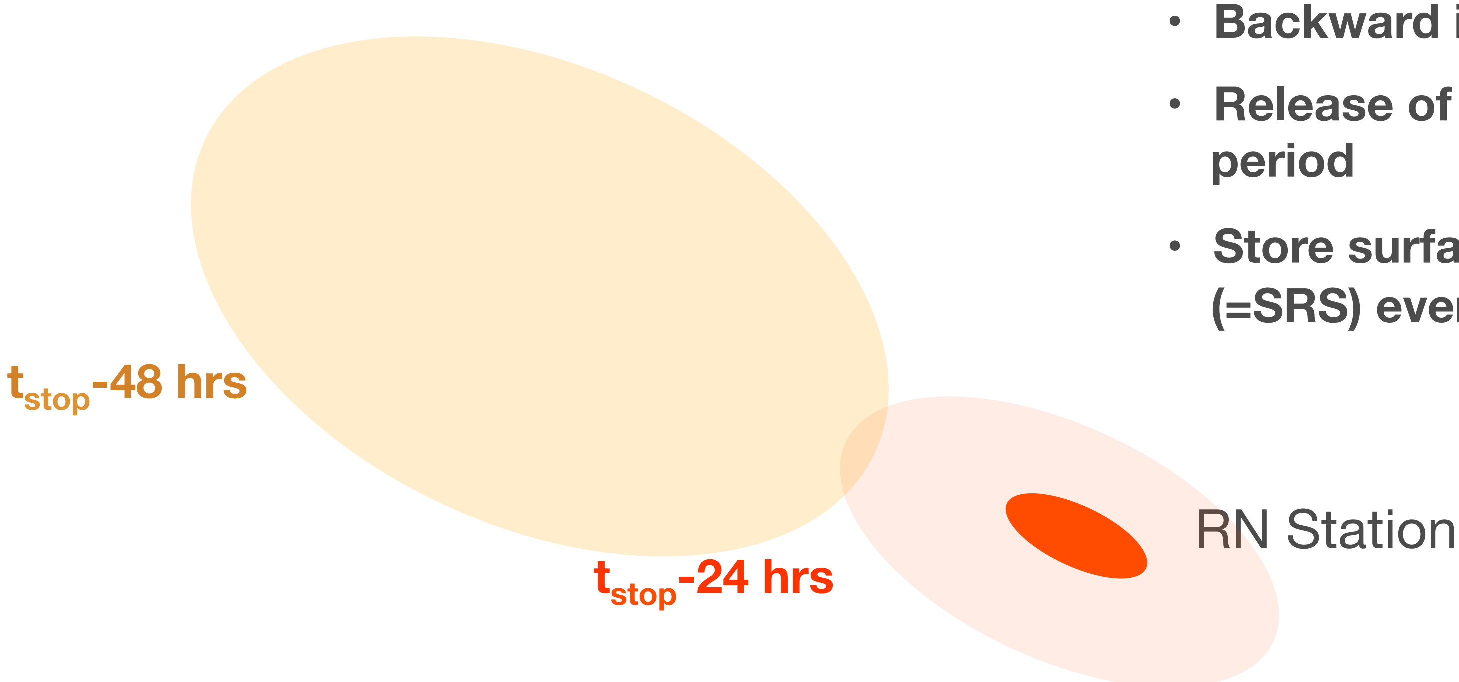

## Sources and Detector

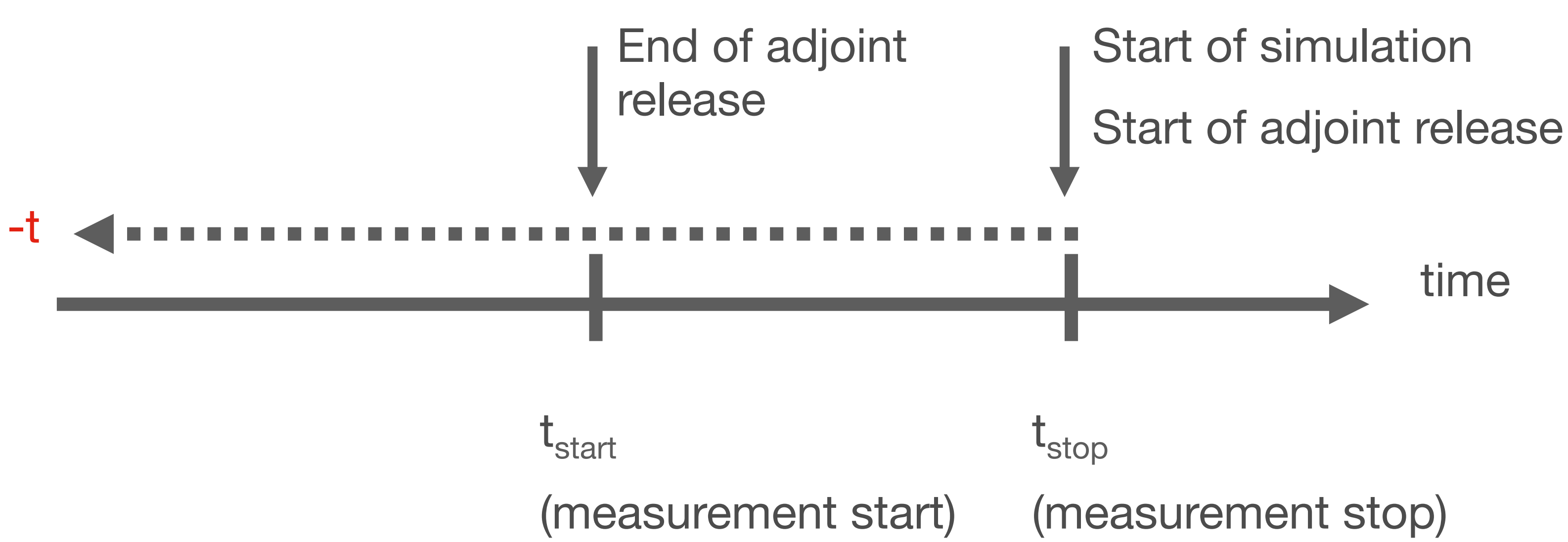



## Atmospheric dynamics responsible for transport

## Understanding of signals at stations

- **Backward in time integration**
- **Release of 10 000 particles/h across 24h-sampling period**
- **Store surface level „retro-plume“ concentration (=SRS) every 3 hours**

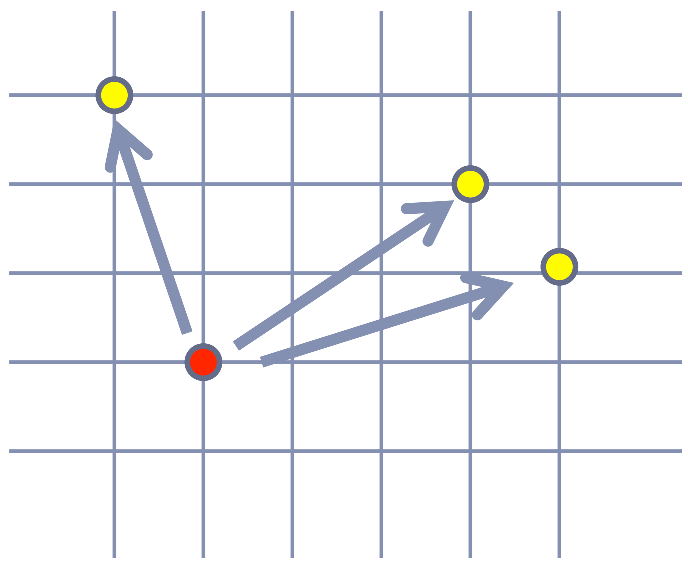


## **Multiple contributions:** $c = \sum M_{i,n} \cdot S_{i,n}$

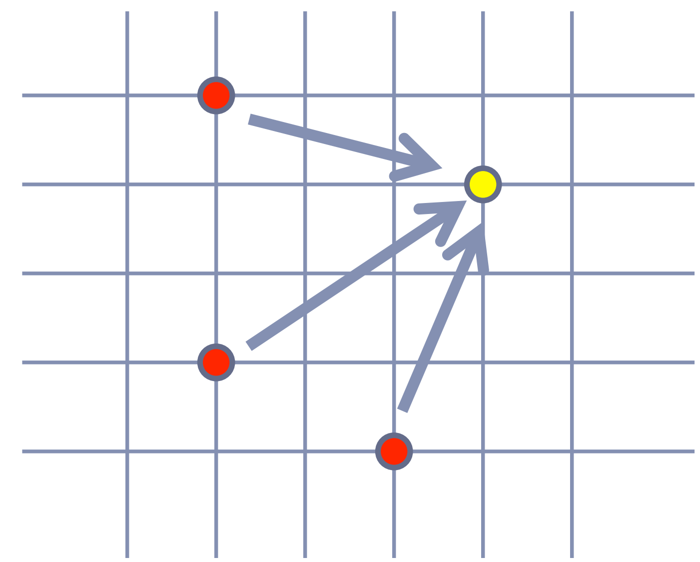
i sources, n time steps

Forward simulations:



One source, multiple receptors

Backward simulations:



Multiple sources, one receptor

- Typical atmospheric dilutions: $10^{-14}$ to $10^{-15}$

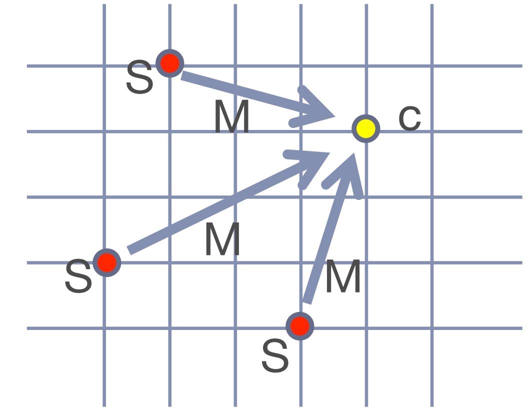
| | | |
|---|---|---|
| Comparison: | Emissions | $10^{10}$-$10^{14}$ Bq |
| | MDC | $10^{-3}$ Bq/m$^3$ |

- Source-Receptor-Sensitivity

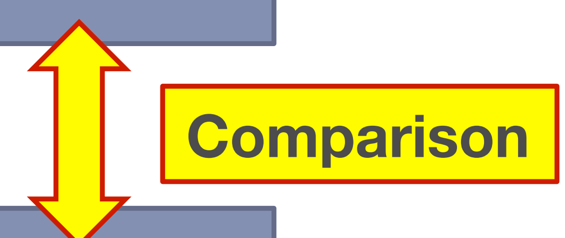
$$c = \sum M_{i,n} \cdot S_{i,n}$$

***S***=source term ***M*** =sensitivity ***c***=concentration

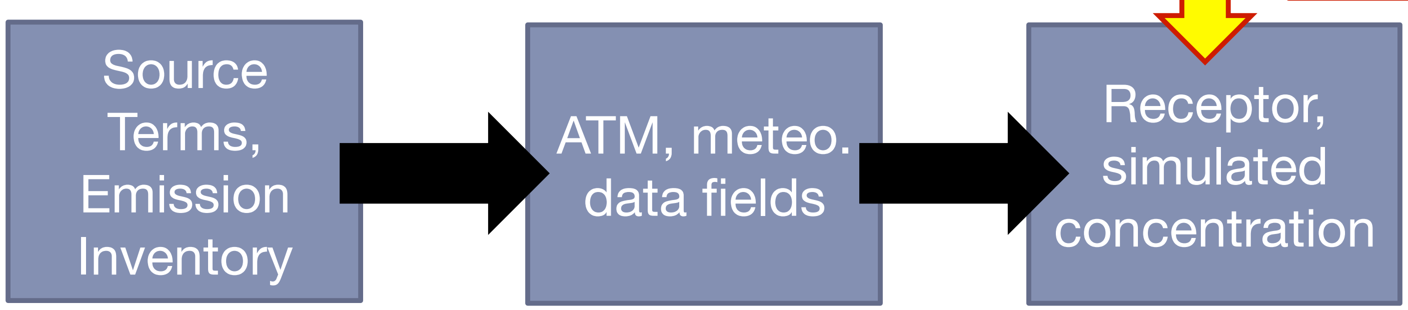
Backward simulations:



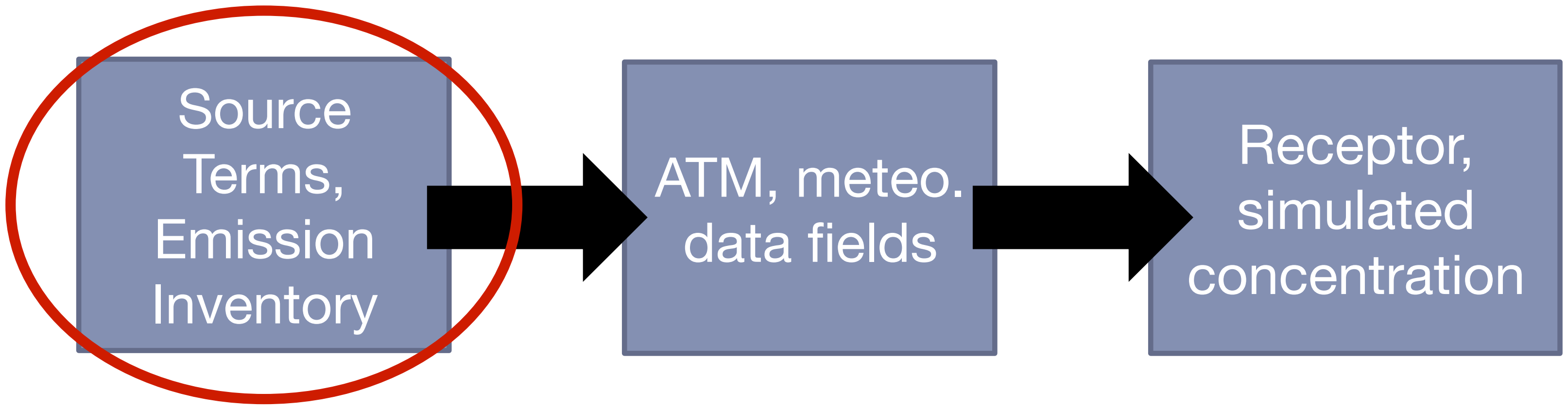

- Reality

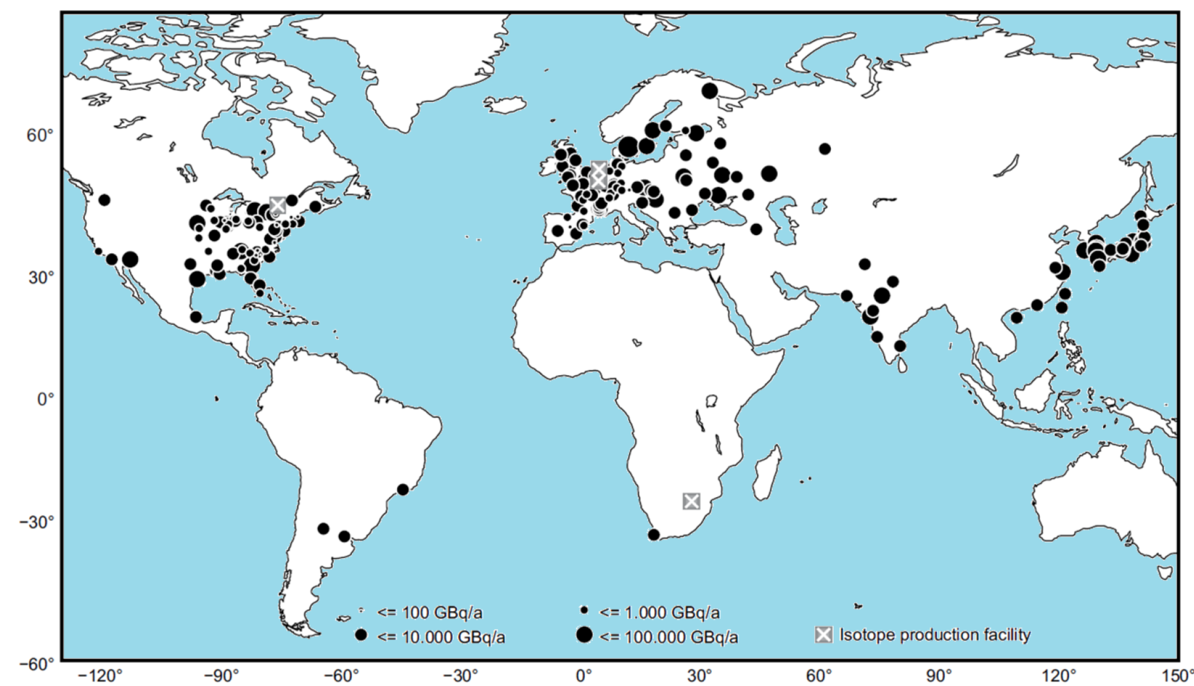

- Simulation




Schoeppner M. and Plastino W., (2014). Determination of the global coverage of the IMS xenon-133 component for the detection of nuclear explosions, Science & Global Security, 22, 209-234.




- Emission inventory
  - Built from publications and personal contact with plant operators
  - Needed: Location (latitude/longitude), and emissions
- Annual averages of estimations from:
  - 200 nuclear power plants
  - 5 isotope production facilities




Schoeppner M. and Plastino W., (2014). Determination of the global coverage of the IMS xenon-133 component for the detection of nuclear explosions, Science & Global Security, 22, 209-234.

| Country | Site Name | Lat | Lon | $^{133}$Xe GBq/a |
|---|---|---|---|---|
| Australia | ANSTO (Sydney) | -34.0 | 151.0 | 5.00E+05 |
| Belgium | IRE (Fleurus) | 50.0 | 4.0 | 1.00E+06 |
| Canada | CRL (Chalkriver) | 46.0 | -77.0 | 1.00E+07 |
| Netherlands | NRG (Petten) | 53.0 | 5.0 | 7.30E+02 |
| South Africa | NECSA(Pelindaba) | -25.0 | 27.0 | 4.00E+06 |

| Country | Site Name | Lat | Lon | $^{131m}$Xe | $^{133m}$Xe | $^{133}$Xe | $^{135}$Xe GBq/a |
|---|---|---|---|---|---|---|---|
| Argentina | Atucha | -34.0 | -59.0 | 1.09E+02 | 3.42E+01 | 1.24E+03 | 8.97E+02 |
| | Embalse | -32.0 | -64.0 | 1.09E+02 | 3.42E+01 | 1.24E+03 | 8.97E+02 |
| Armenia | Armenia | 40.0 | 44.0 | 1.09E+02 | 3.42E+01 | 1.24E+03 | 8.97E+02 |
| Belgium | Doel | 51.0 | 4.0 | 4.36E+02 | 1.37E+02 | 4.97E+03 | 3.59E+03 |
| | Tihange | 50.0 | 5.0 | 3.27E+02 | 1.03E+02 | 3.72E+03 | 2.69E+03 |
| Brazil | Angra | -23.0 | -44.0 | 2.18E+02 | 6.84E+01 | 2.48E+03 | 1.79E+03 |
| Bulgaria | Kozloduy | 43.0 | 23.0 | 2.18E+02 | 6.84E+01 | 2.48E+03 | 1.79E+03 |
| Canada | Bruce | 44.0 | -81.0 | 6.54E+02 | 2.05E+02 | 7.45E+03 | 5.38E+03 |

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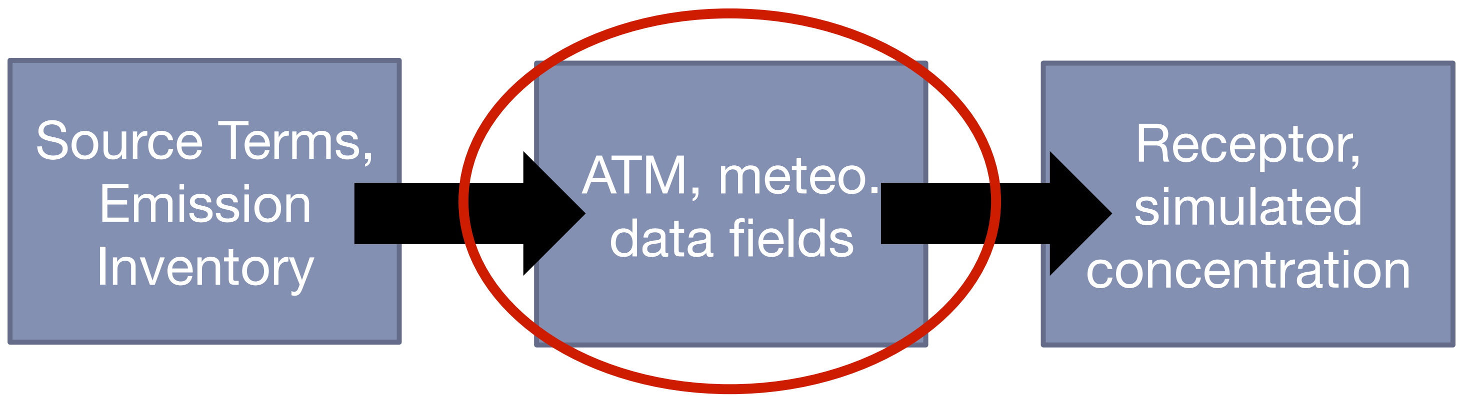
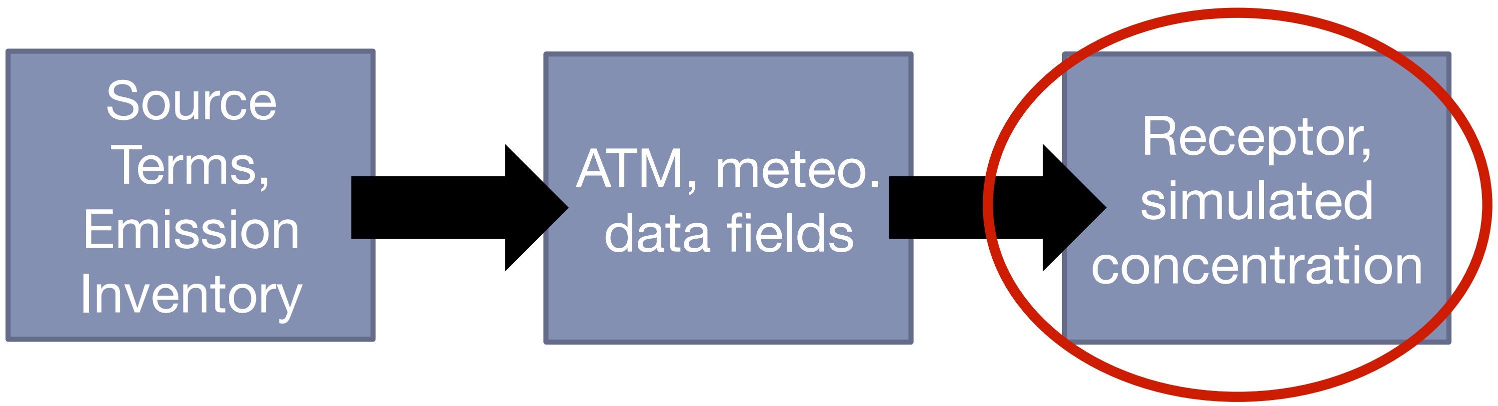
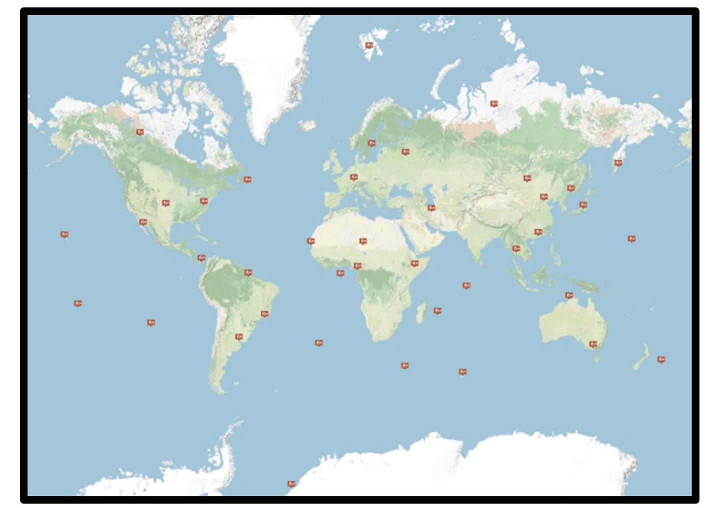



- Meteorological data:
  - 1° x 1° Latitude/Longitude spatial resolution
  - 3h time resolution
  - Storage and management of ca. 0.5 GB per day of data

  - Supplied by the European Centre for Medium-Range Weather Forecasts

Schoeppner M. and Plastino W., (2014). Determination of the global coverage of the IMS xenon-133 component for the detection of nuclear explosions, Science & Global Security, 22, 209-234.



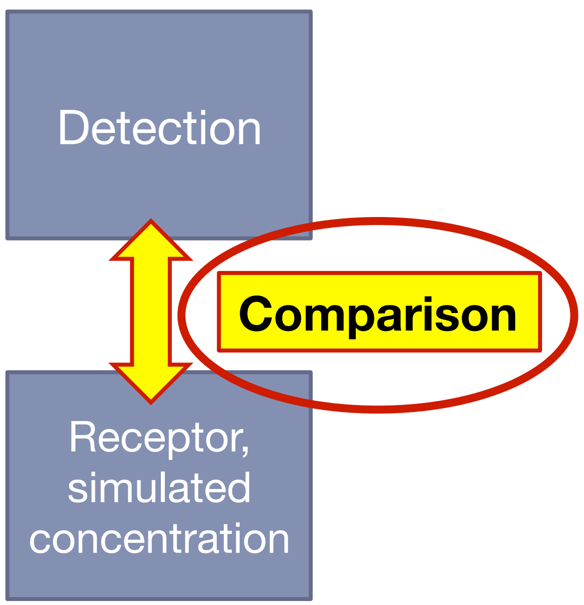
- Conducted simulations:
  - Annual time series for 2010
  - for 40 stations (12h/24h sampling time)
  - 14 weeks backward simulation time
  - Ca. 22.000 simulations,  each ca. 4 hours of CPU-time

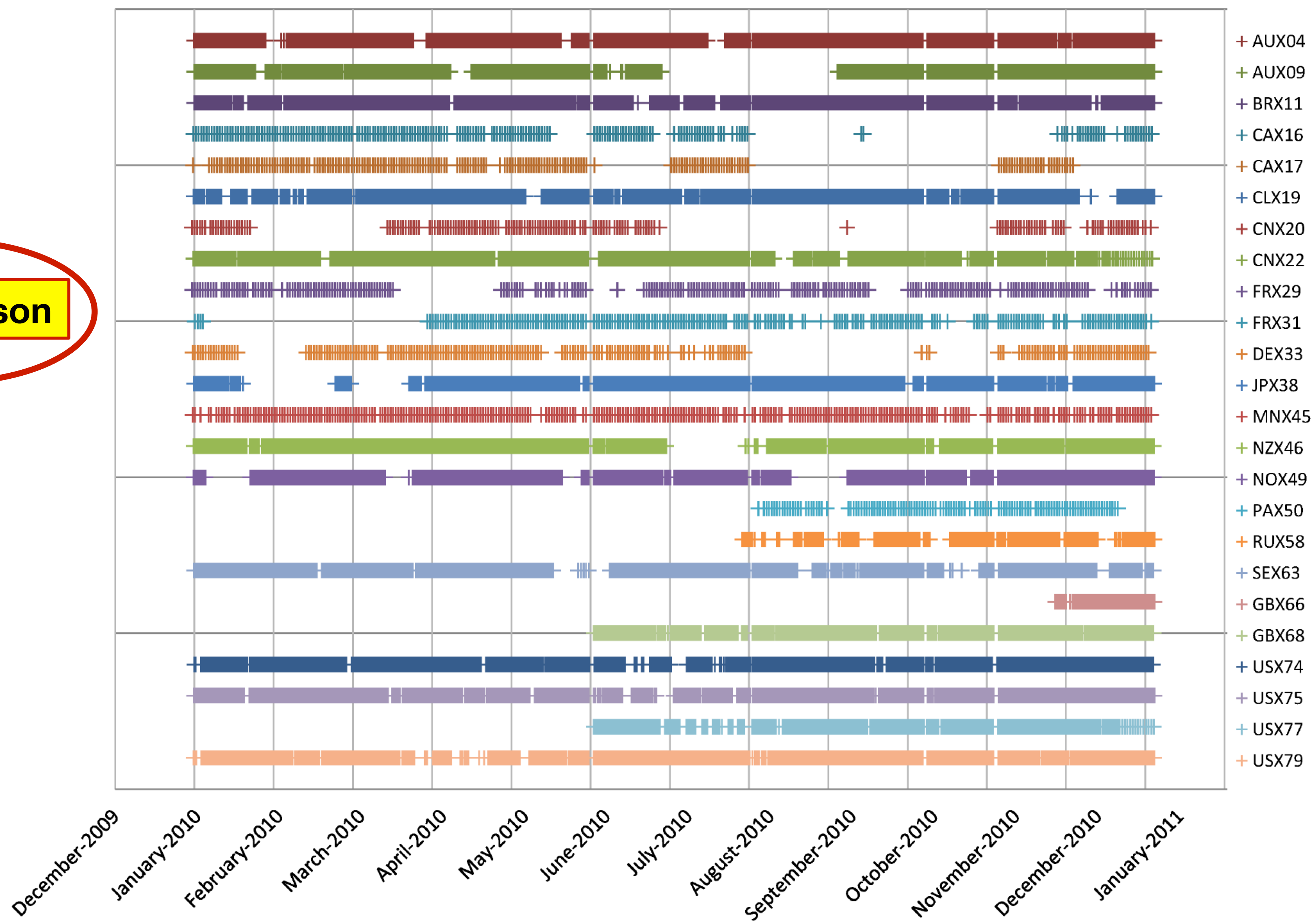
- 40 station codes

  Country+Xenon+No:
  - AUX04
  - DEX33
  - USX74
  - …

Schoeppner M. and Plastino W., (2014). Determination of the global coverage of the IMS xenon-133 component for the detection of nuclear explosions, Science & Global Security, 22, 209-234.

- Available experimental data from 2010

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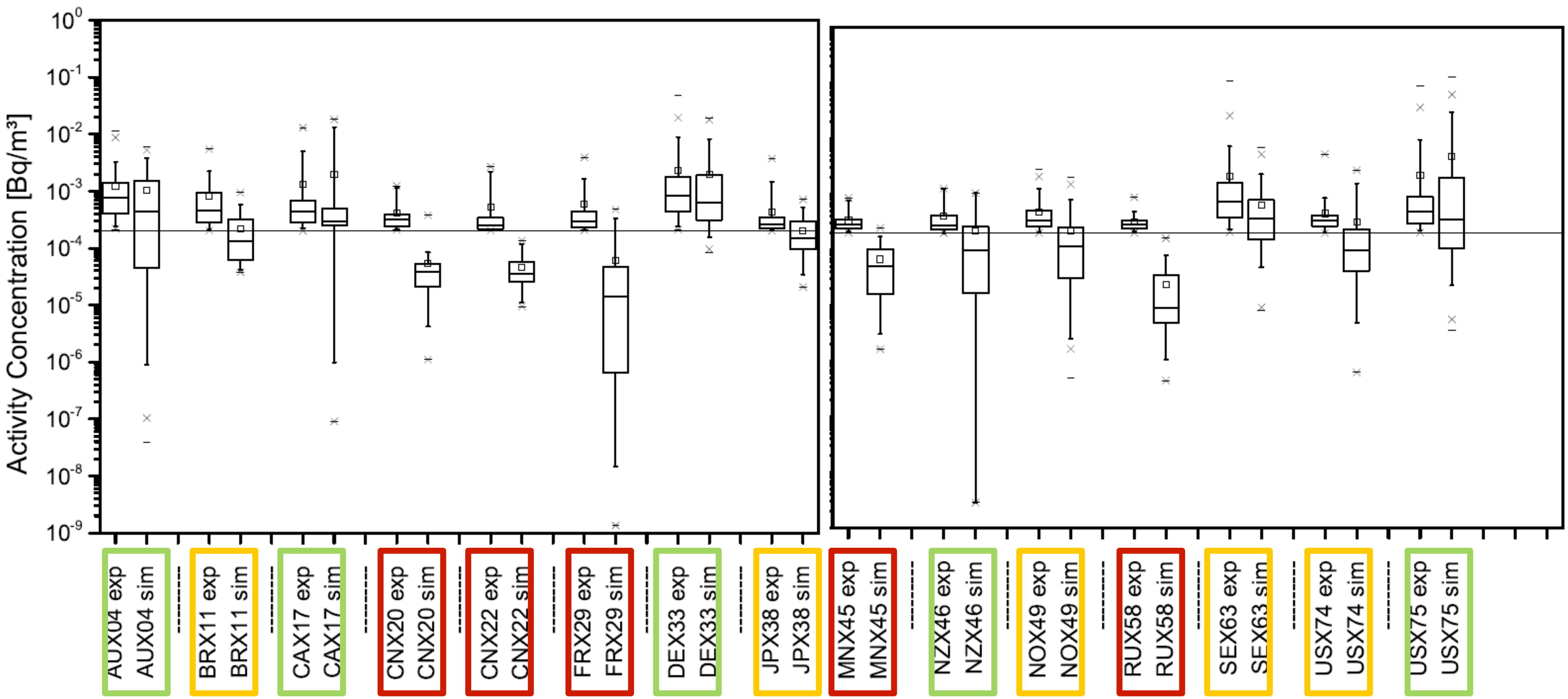
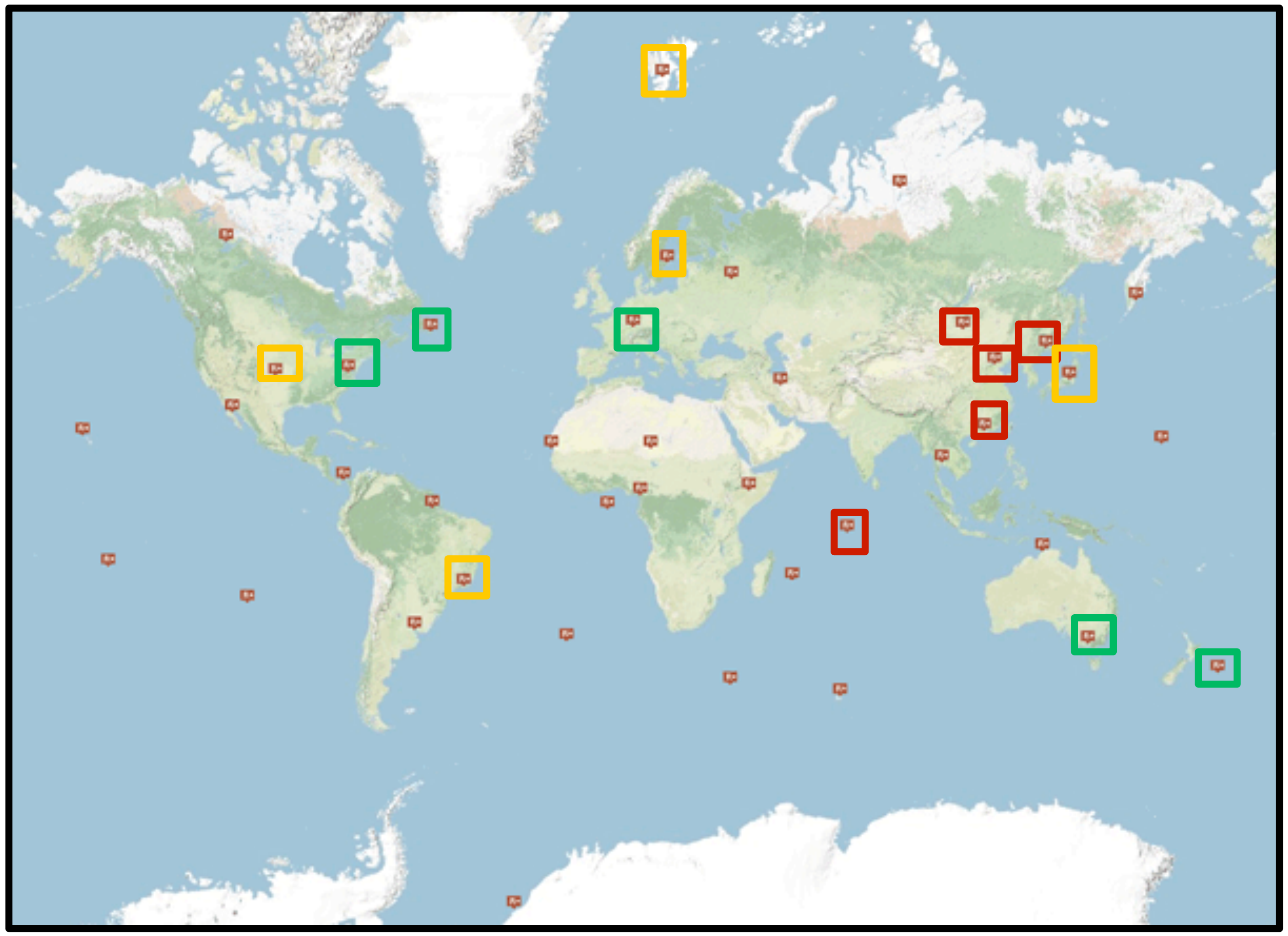



Schoeppner M. and Plastino W., (2014). Determination of the global coverage of the IMS xenon-133 component for the detection of nuclear explosions, Science & Global Security, 22, 209-234.

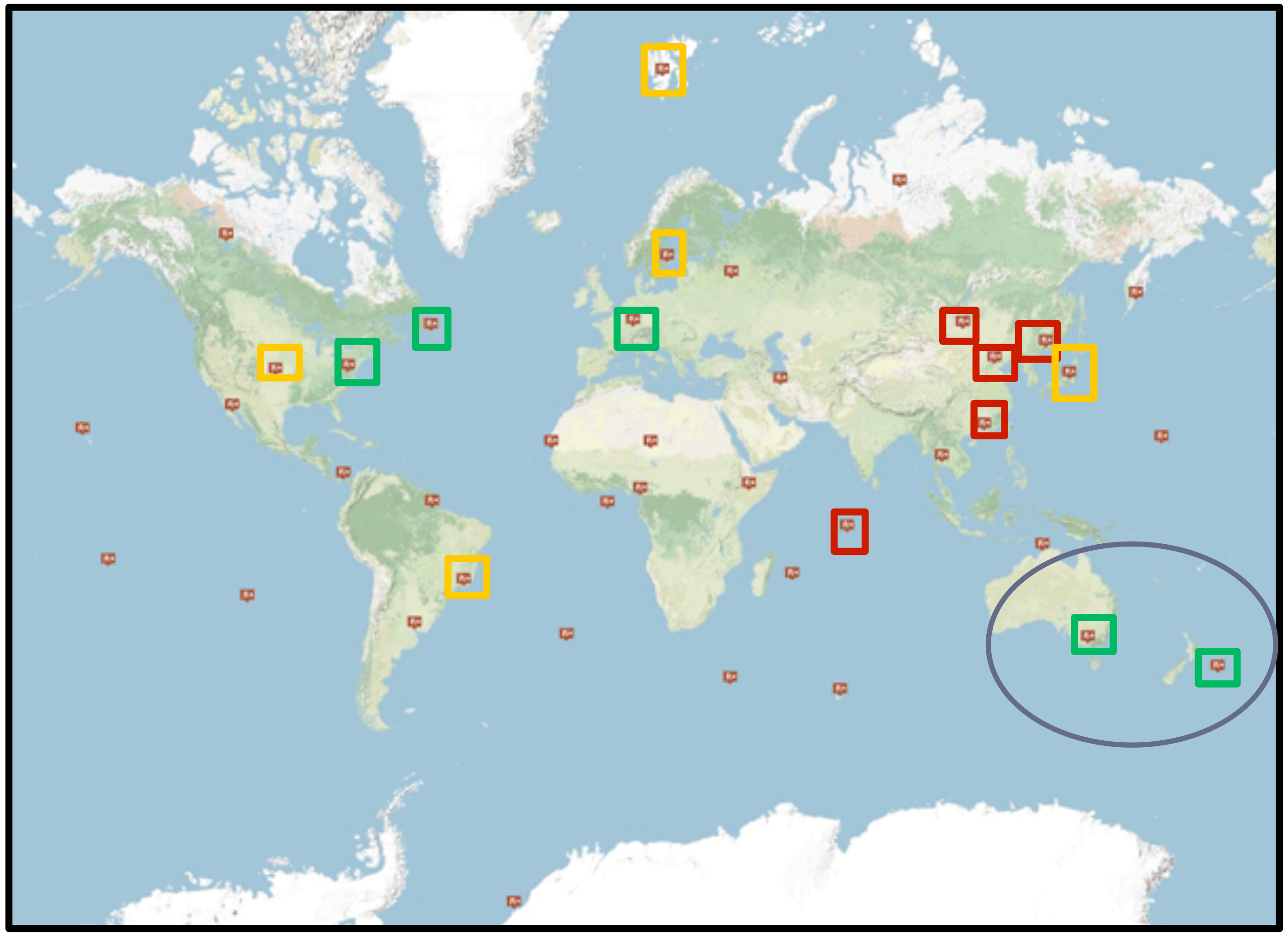
- Comparison of simulations with experimental data



- **Regional dependence of under-estimation**

Schoeppner M. and Plastino W., (2014). Determination of the global coverage of the IMS xenon-133 component for the detection of nuclear explosions, Science & Global Security, 22, 209-234.

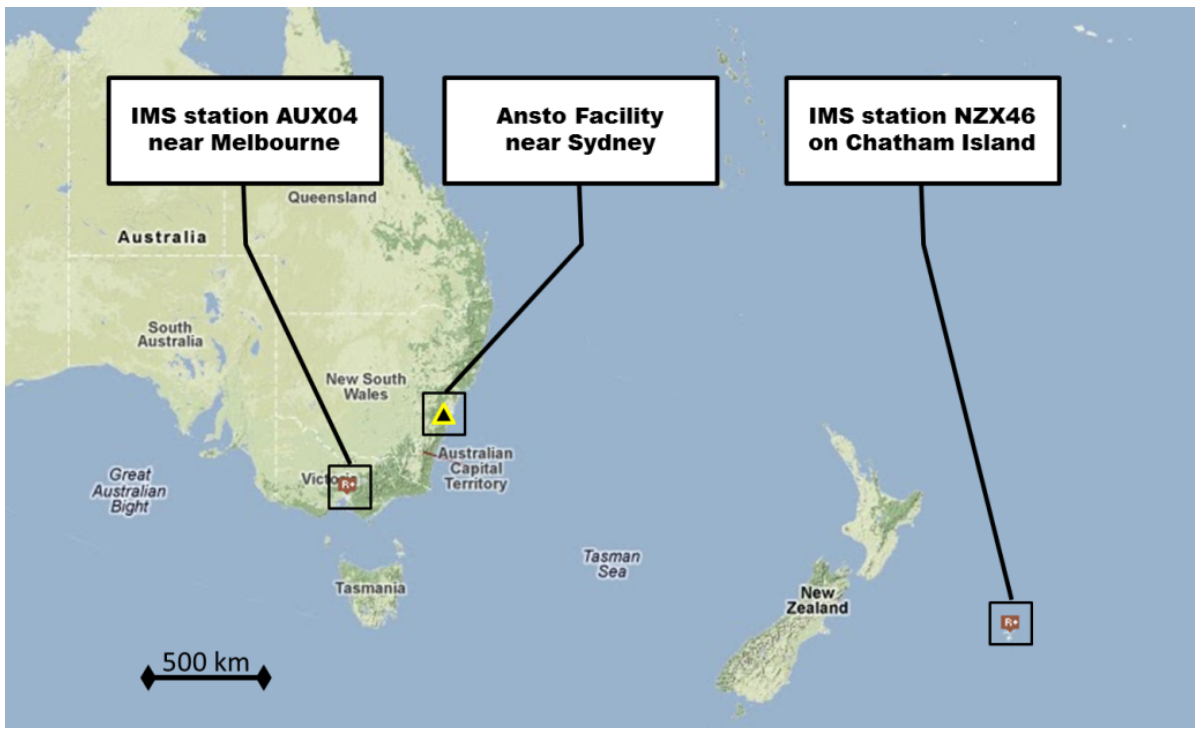
- Comparison of simulations with experimental data



- **Regional dependence of under-estimation**

Schoeppner M. and Plastino W., (2014). Determination of the global coverage of the IMS xenon-133 component for the detection of nuclear explosions, Science & Global Security, 22, 209-234.

- Atmospheric radioxenon situation in Oceania




- Emission data for $^{133}Xe$ with daily resolution provided from ANSTO facility
  - 4 data sets created with emission with various time resolution
    - Daily
    - Weekly
    - Monthly
    - Annual

What is the influence of different time resolutions of emission data?

Schoeppner M., Plastino W., Hermanspahn N., Hoffmann E., Kalinowski M., Orr B., Tinker R. (2013), Atmospheric transport modelling of time resolved $^{133}Xe$ emissions from the isotope production facility ANSTO, Australia, Journal of Environmental Radioactivity, 126, 1-7.

The statistical results of the time series analysis for both IMS stations and the four data sets.

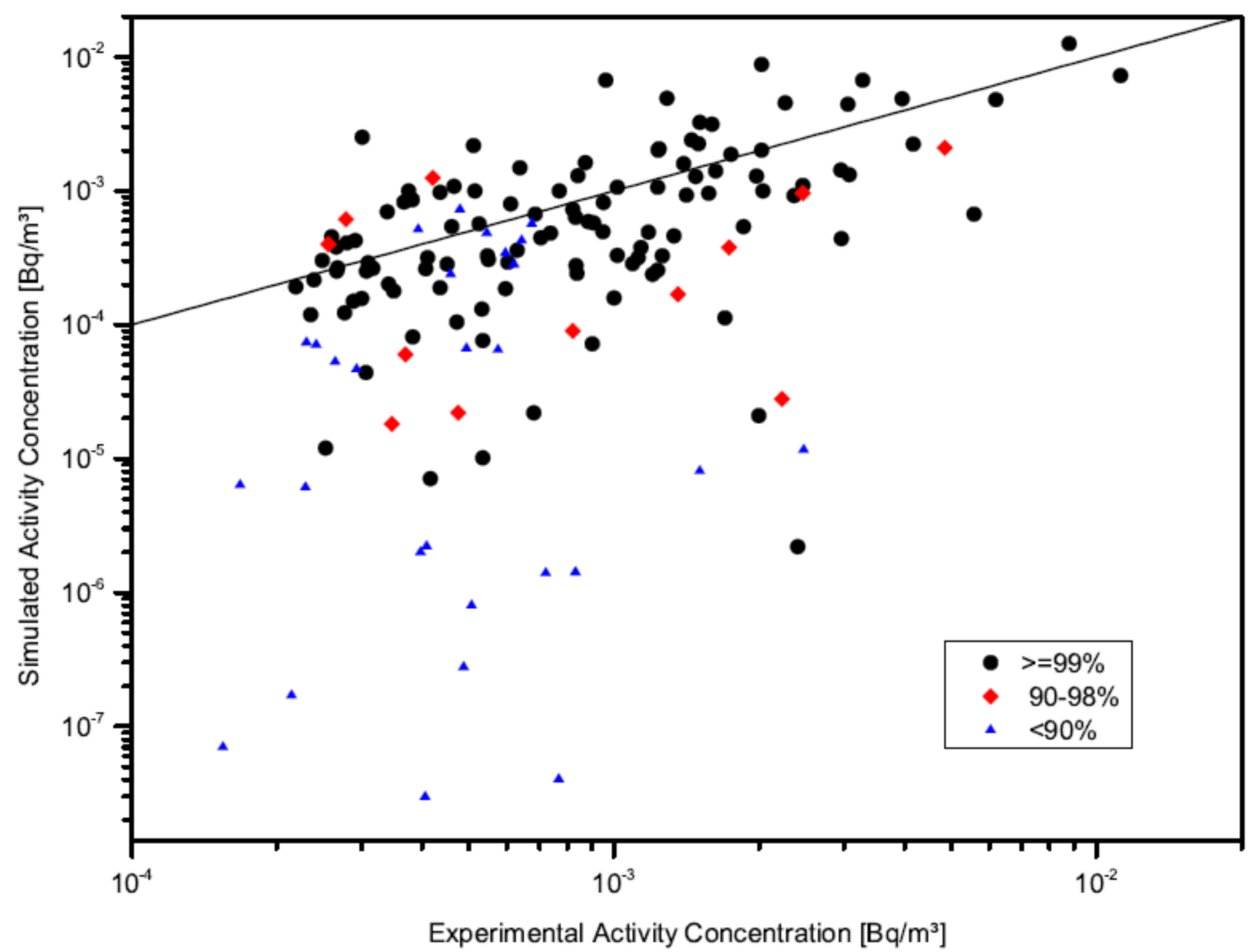
| | | ANSTO source term time resolution | | | |
|---|---|---|---|---|---|
| Station | Parameter | Daily | Weekly | Monthly | Annual |
| AUX04 | Correlation | 0.67 | 0.49 | 0.47 | 0.39 |
| | NMSE | 1.13 | 1.45 | 1.42 | 1.63 |
| | Fractional bias | 0.16 | 0.12 | 0.23 | 0.15 |
| NZX46 | Correlation | 0.06 (0.92) | 0.04 (0.77) | 0.02 (0.76) | −0.02 (0.73) |
| | NMSE | 0.20 (0.01) | 0.20 (0.02) | 0.23 (0.03) | 0.31 (0.04) |
| | Fractional bias | 0.90 (0.41) | 0.86 (0.36) | 0.98 (0.51) | 1.18 (0.75) |

Values in parentheses are for NZX-46 excluding one under-predicted outlier. NMSE stands for normalized mean square error.

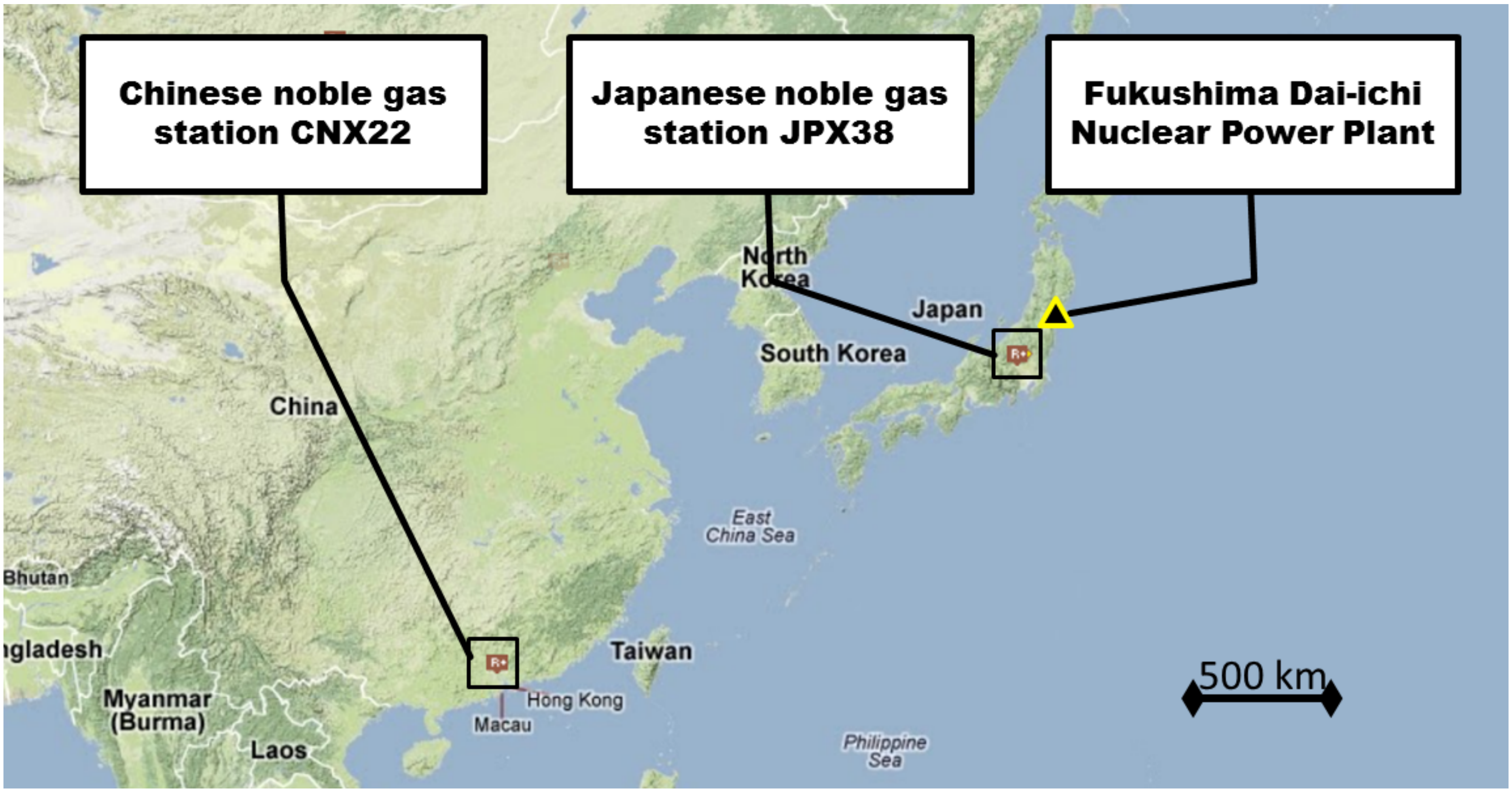
Prediction quality of data subsets sorted after share from the ANSTO facility.

| | Pearson correlation coefficient/fractional bias | | | |
|---|---|---|---|---|
| | ANSTO source term time resolution | | | |
| Share[a] | Daily | Weekly | Monthly | Annual |
| >=99% | 0.68/0.03 | 0.48/−0.02 | 0.47/0.11 | 0.41/−0.03 |
| 90−98% | 0.67/0.88 | 0.55/0.92 | −0.05/0.82 | −0.09/0.97 |
| <90% | −0.04/1.19 | −0.04/1.32 | −0.08/1.27 | −0.07/1.25 |

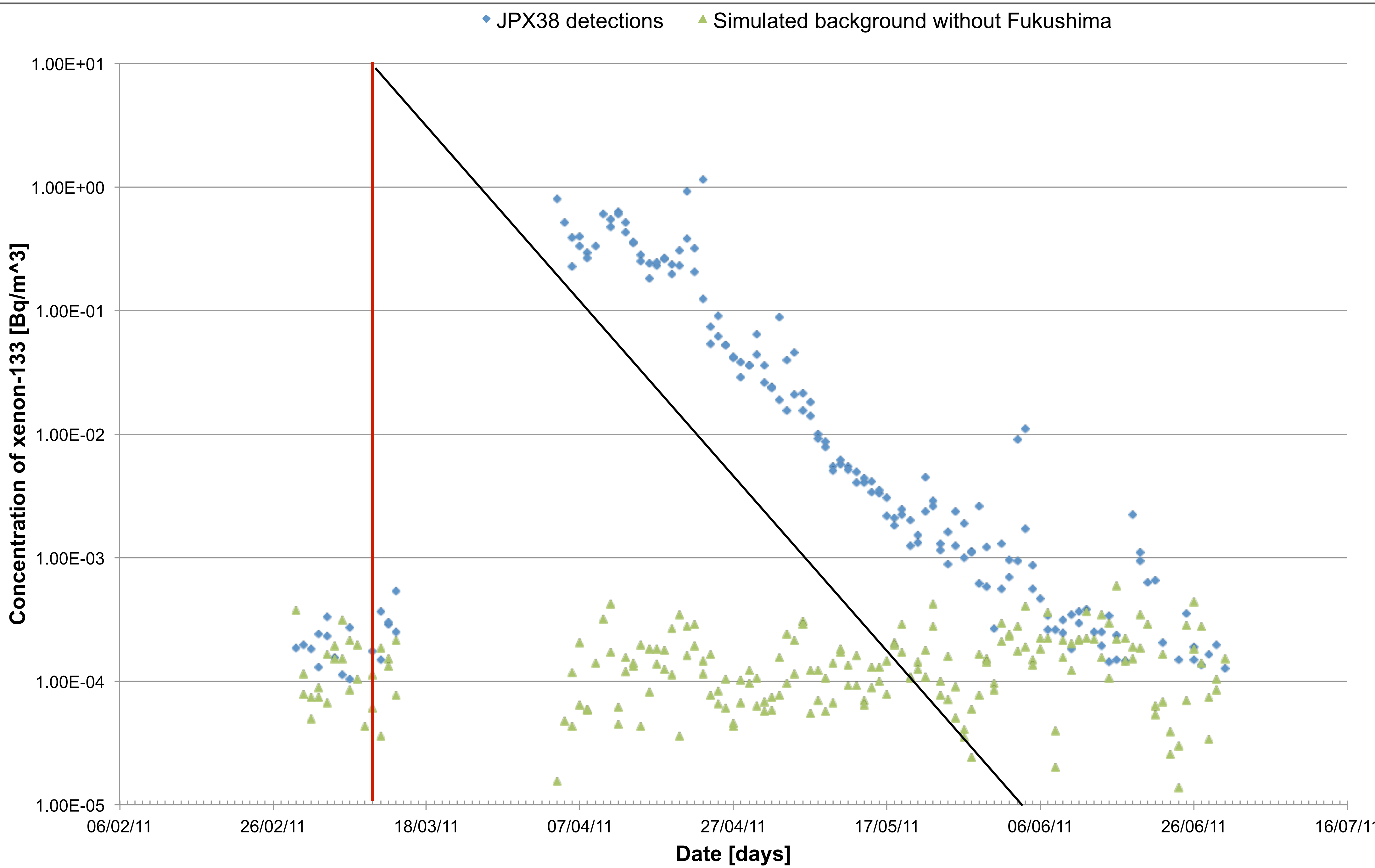
[a] Contribution of ANSTO to each simulated concentration.



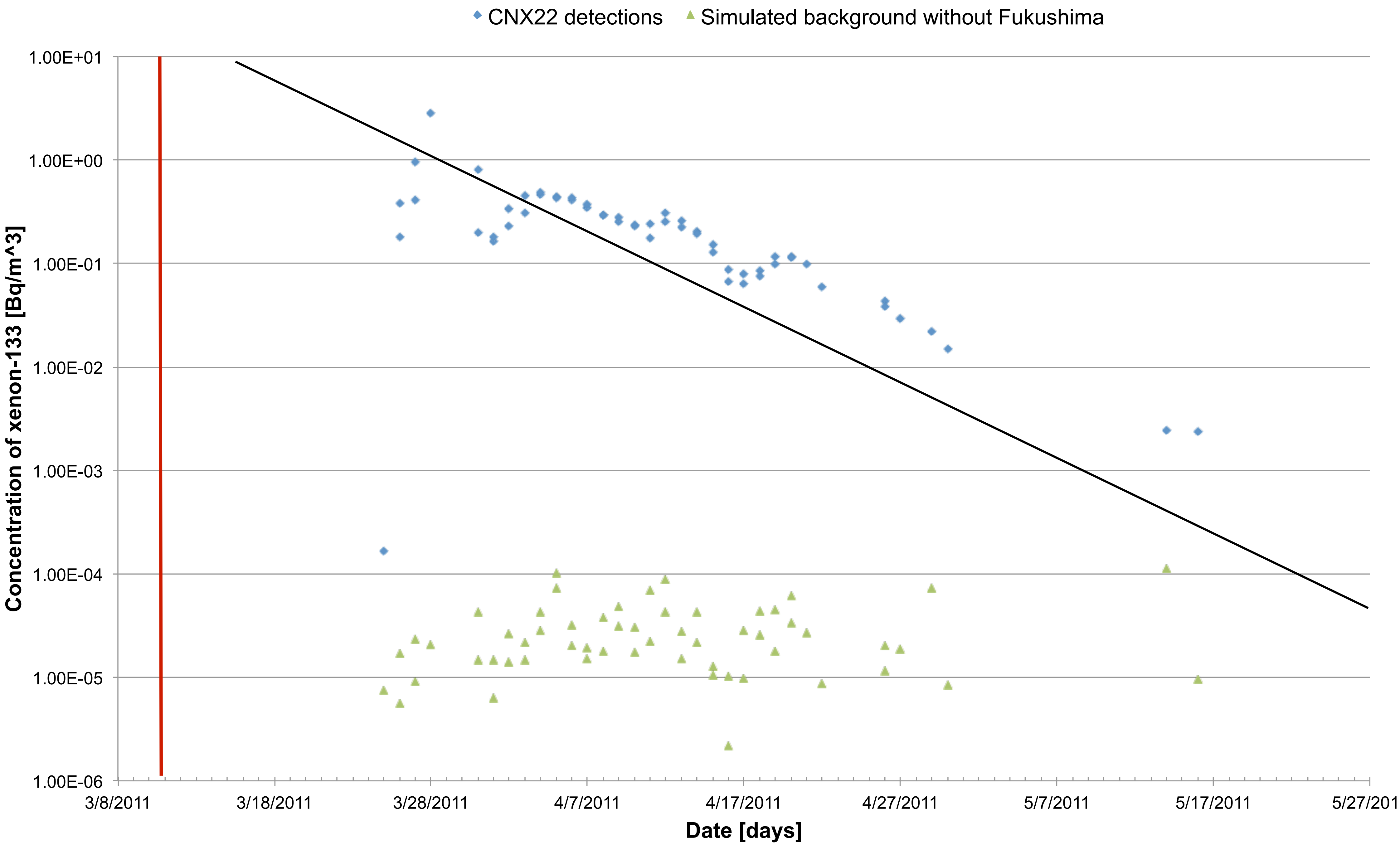

Schoeppner M., Plastino W., Hermanspahn N., Hoffmann E., Kalinowski M., Orr B., Tinker R. (2013), Atmospheric transport modelling of time resolved $^{133}Xe$ emissions from the isotope production facility ANSTO, Australia, Journal of Environmental Radioactivity, 126, 1-7.




Schoeppner, M., Plastino W., Povinec P., Nikkinen M., *et al.,* (2013) Estimation of the Radioactive Source Dispersion from Fukushima Nuclear Power Plant Accident. Applied Radiation and Isotopes, 81, 358-361.




Schoeppner, M., Plastino W., Povinec P., Nikkinen M., *et al.,* (2013) Estimation of the Radioactive Source Dispersion from Fukushima Nuclear Power Plant Accident. Applied Radiation and Isotopes, 81, 358-361.




Schoeppner, M., Plastino W., Povinec P., Nikkinen M., *et al.,* (2013) Estimation of the Radioactive Source Dispersion from Fukushima Nuclear Power Plant Accident. Applied Radiation and Isotopes, 81, 358-361.

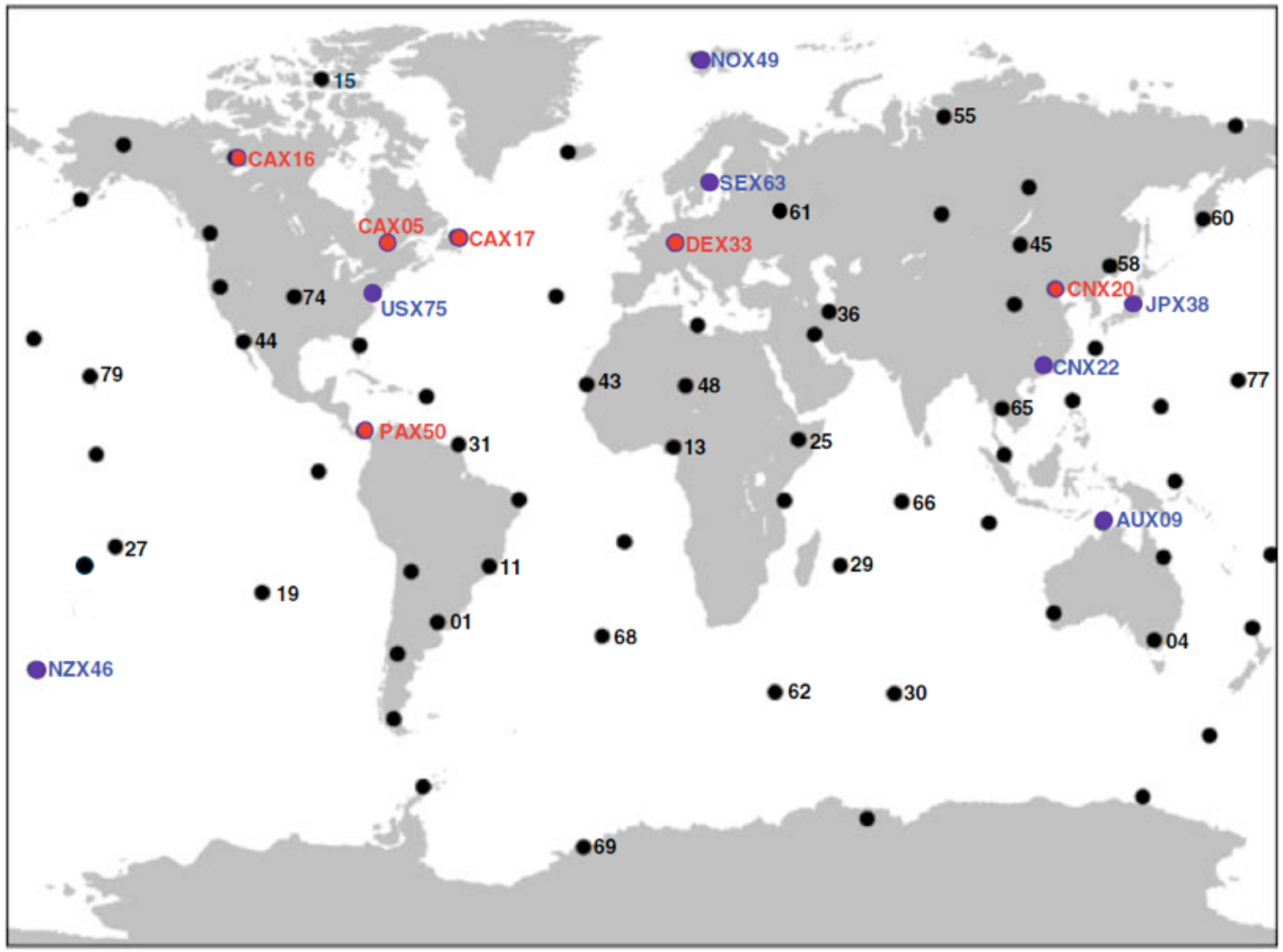



Figure 1
Map showing the location of all the active SPALAX and SAUNA stations in the INGE network in June 2008. The *colour codes* indicate the system type: *red*—SPALAX, *blue*—SAUNA. All other scheduled INGE network locations are indicated by *numbered black dots*. *Unnumbered black dots* indicate IMS RN stations scheduled to have no noble gas measurement capability (colour figure online)

Plastino, W. et al. (2010). Radioxenon Time Series and Meteorological Pattern Analysis for CTBT Event Categorisation. Pure and Applied Geophysics 167, 559-573.

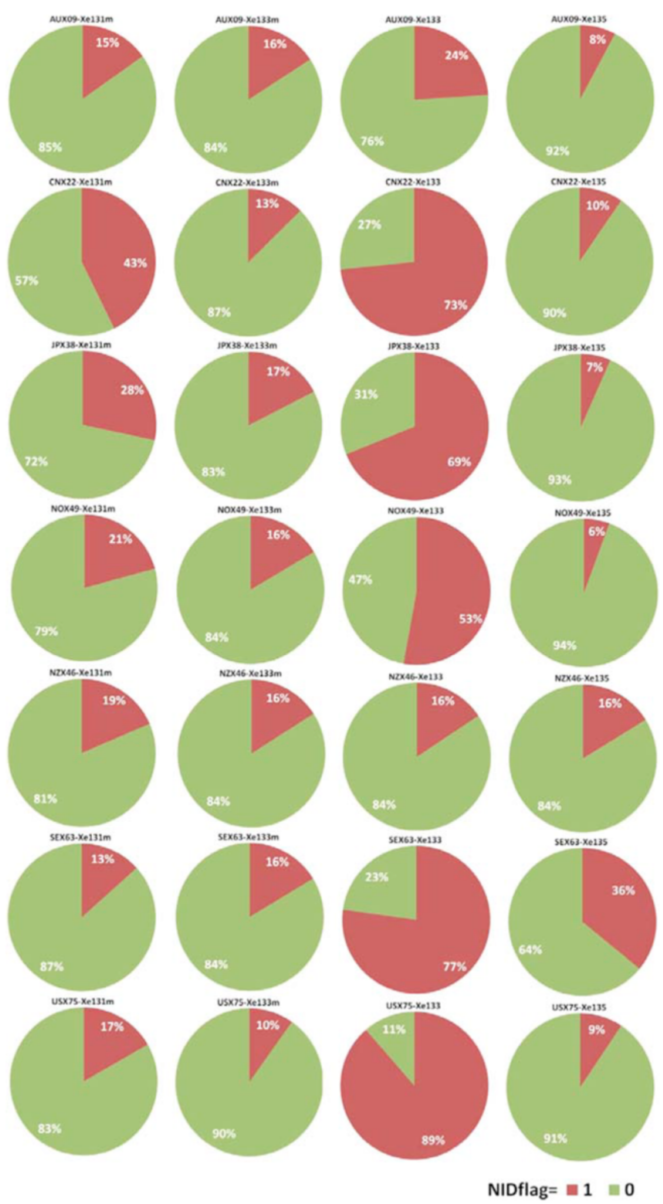



Figure 4
Radioxenon isotopes ($^{131m}Xe$, $^{133m}Xe$, $^{133}Xe$, and $^{135}Xe$) presence by NIDflag codes (*NID* nuclide identification; *1*—nuclide found above critical limit at key line position, *0*—no nuclide found above critical limit at key line position) of the INGE noble gas stations SAUNA comprising series 1 and 2 (Fig. 2): AUX09 (Darwin, Australia), CNX22 (Guangzhou, China), JPX38 (Takasaki-Gunma, Japan), NOX49 (Spitsbergen, Norway), NZX46 (Chatham Island, New Zealand), SEX63 (Stockholm, Sweden), USX75 (Charlottesville, USA)

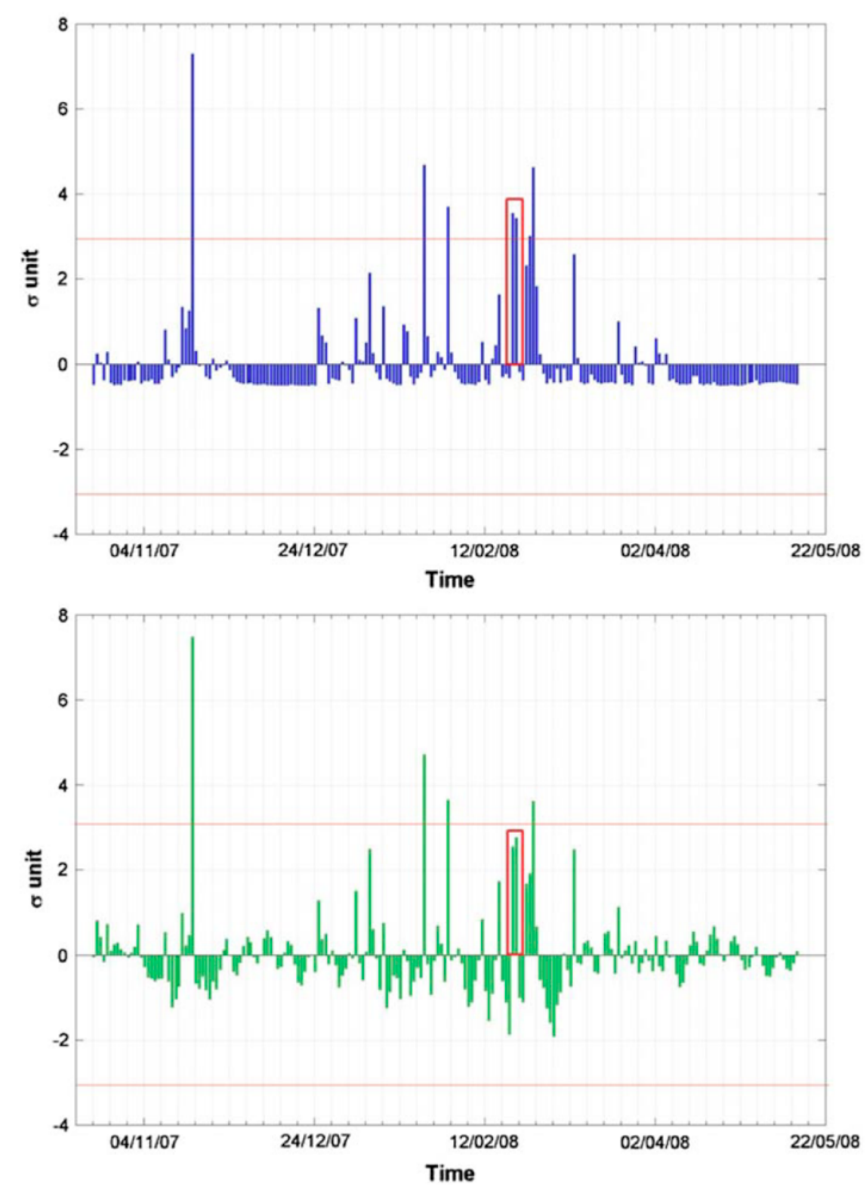
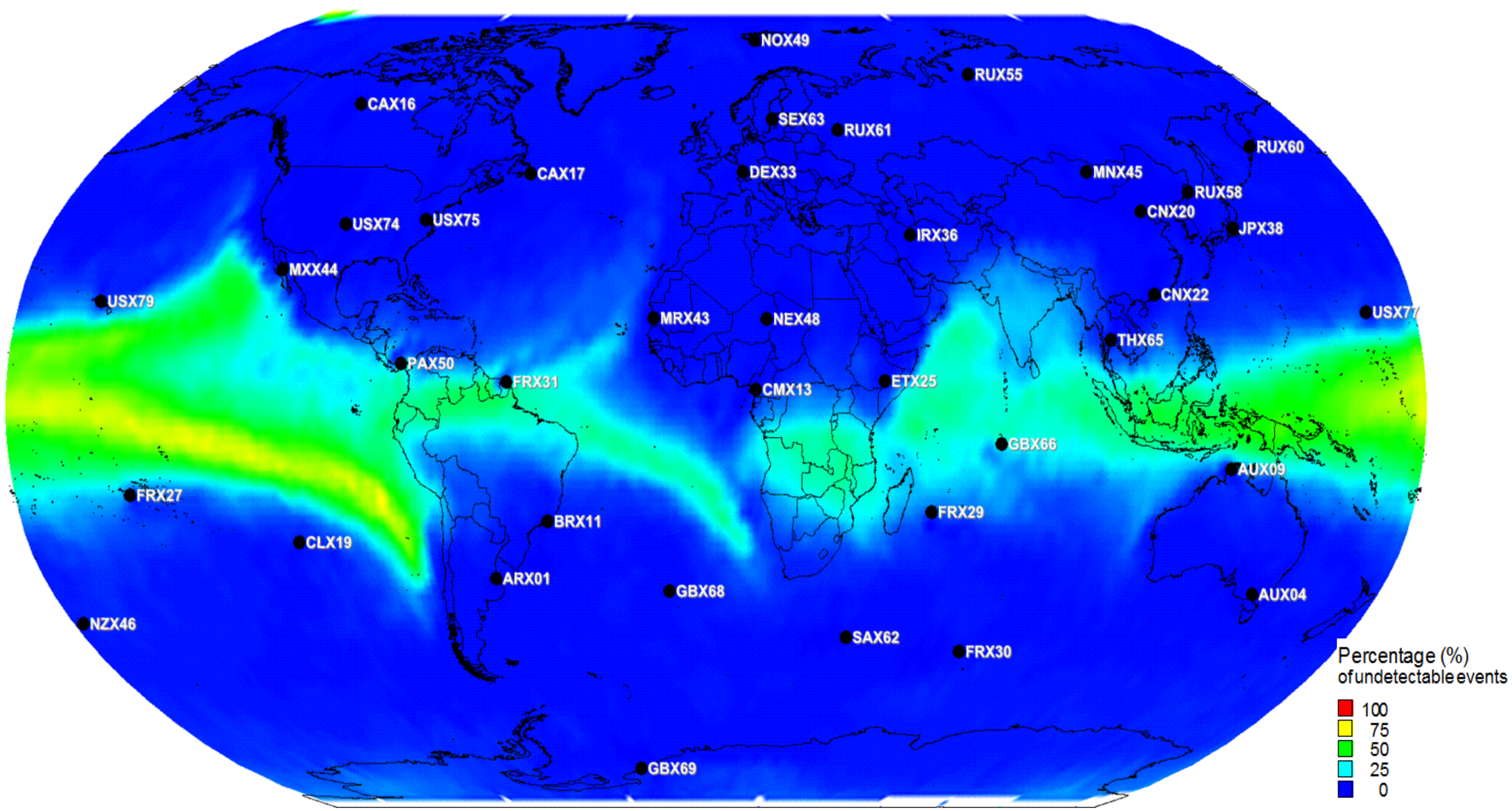



Figure 10
The original (*top*) and residual (*bottom*) time series normalised to null mean value and unit standard deviation for the INGE noble gas station located in St. John's, Canada. The estimated 3$\sigma$, i.e., the thresholds used to identify the outliers, are also shown. The *red rectangles* show the different occurrence probability for the same outlier in the original and residual time series (colour figure online)

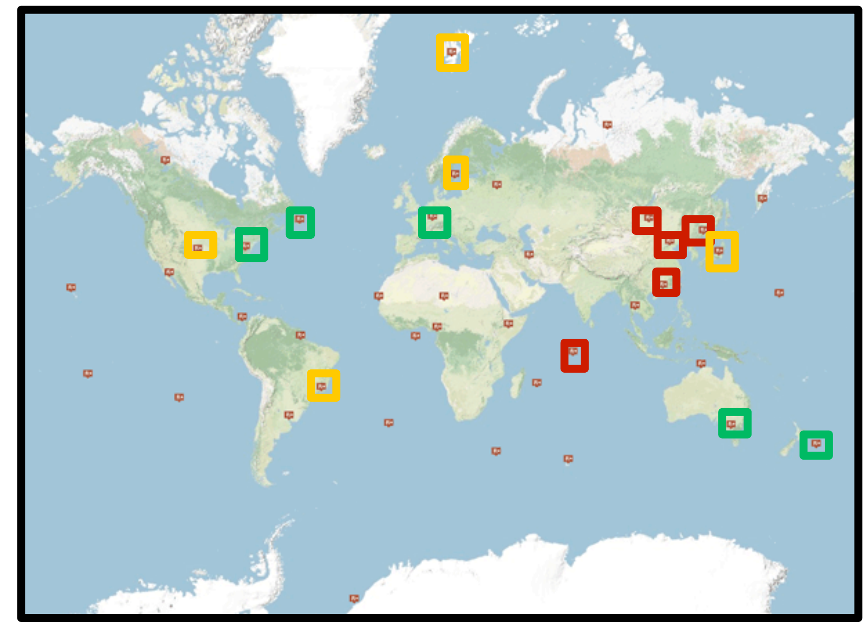
Plastino, W. et al. (2010). Radioxenon Time Series and Meteorological Pattern Analysis for CTBT Event Categorisation. Pure and Applied Geophysics 167, 559-573.

- Undetectable events:
  - Events that do not reach any detector within the simulation time of two weeks



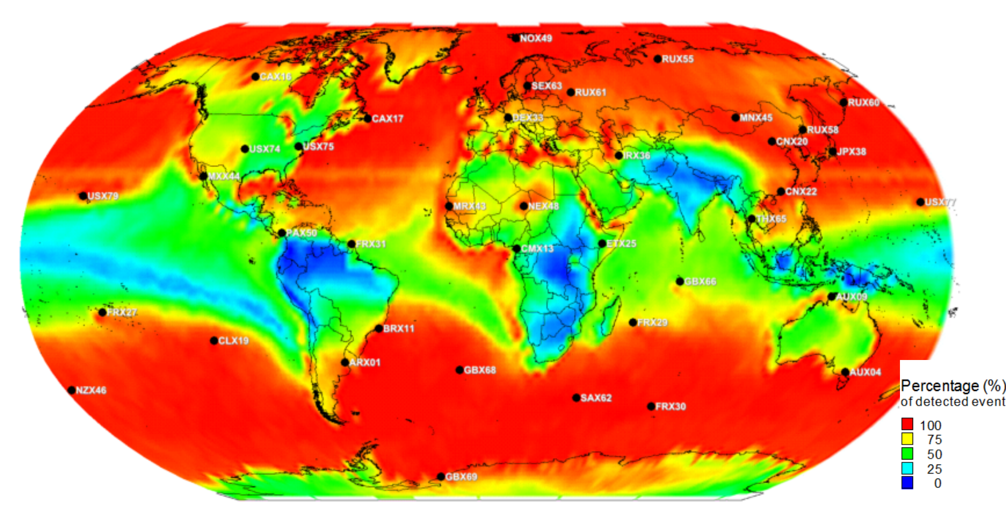

Schoeppner M. and Plastino W., (2014). Determination of the global coverage of the IMS xenon-133 component for the detection of nuclear explosions, Science & Global Security, 22, 209-234.

# Conclusions



- Background generally well understood
  - With regional dependence
    - Europe, America and Oceania well understood
    - Asia clearly underestimated
  - Emission data with time resolution would greatly improve ATM capabilities

- Global coverage of 90% not achieved by current network of stations
  - Only **76.4%** coverage due to
    1. Global background
    2. <u>Undetectable events</u>
  - Improvement possible by
    - Reduction of global background
    - Lower MDC
    - Sophisticated detection criteria
    - **More stations**



Back to the Future