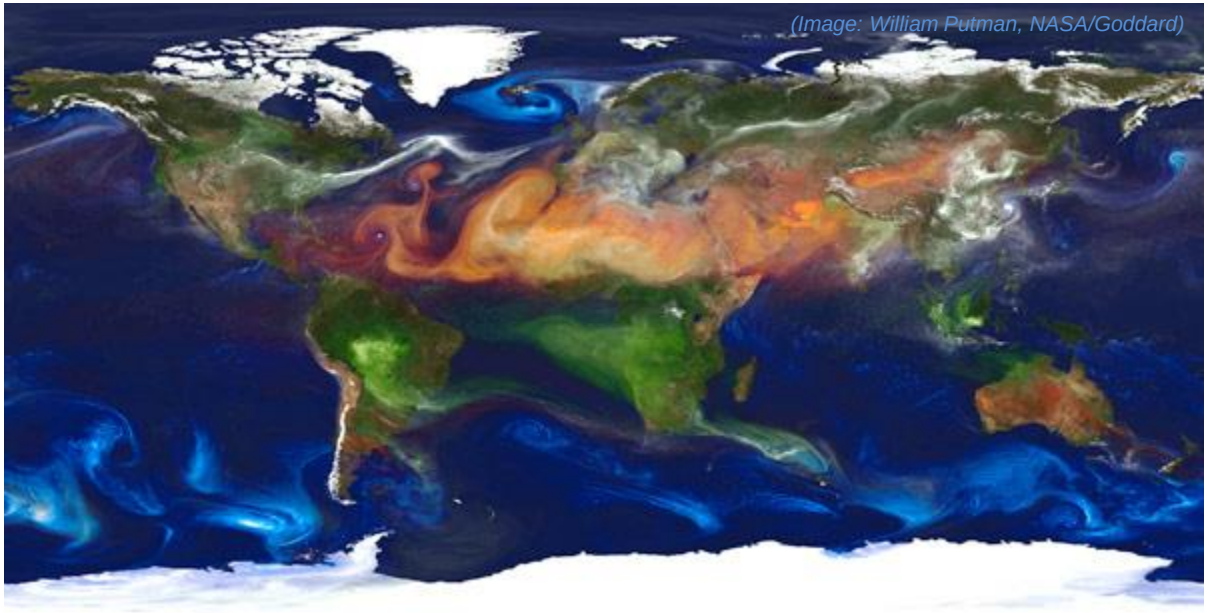


Evaluation of observed and modelled aerosol lifetimes

-using radioactive tracers of opportunity and an ensemble of 19 global models

N. I. Kristiansen[1], A. Stohl[1], T. Christoudias[2], D. Kunkel[3], B. Croft[4] , J. Pierce[4], R. Martin[4], T. Bergman[5], H. Kokkola[5], Y. H. Lee[6], D. Shindell[16], G. Pitari[7], G. Di Genova[7], H. Zhang[8], S. Zhao[8], O. A. Søvde[9], H. Wang[10], R. Easter[10], P-L. Ma[10], P. Rasch[10], K. Zhang[10], X. Liu[11], N. Evangeliou[12], Y. Balkanski[12], K. Tsigaridis[13], S. Bauer[13], H. Klein[14], S. Leadbetter[15], D. J. L., Olivié[14], M. Schulz[14]

1: NILU-Norwegian Institute for Air Research, Kjeller, Norway; 2: Cyprus Institute; 3: Institute for Atmospheric Physics, Johannes Gutenberg-University Mainz, Germany; 4: Department of Physics and Atmospheric Science Dalhousie University; 5: Finish Meteorological Institute; 6: NASA Goddard Institue for Space studies, New York; 7: University of L'Aquila, Italy; 8: Chinese academy of meteorological science; 9: Center for International Climate and Environmental Research – Oslo (CICERO), Oslo, Norway; 10: Pacific Northwest National Laboratory (PNNL), Richland, WA, USA; 11: University of Wyoming; 12: Laboratoire des Sciences du Climat et de l'Environnement, CEA-CNRS-UVSQ, Gif-sur-Yvette, France; 13: NASA Goddard Institute for Space Studies and Columbia Earth Institute, New York, NY, USA; 14: Norwegian meteorological institute; 15: Met Office, Exeter, UK; 16: Earth and Ocean Sciences, Nicholas School of the Environment, Duke University, Durham, NC 27708

AEROCOM-P3

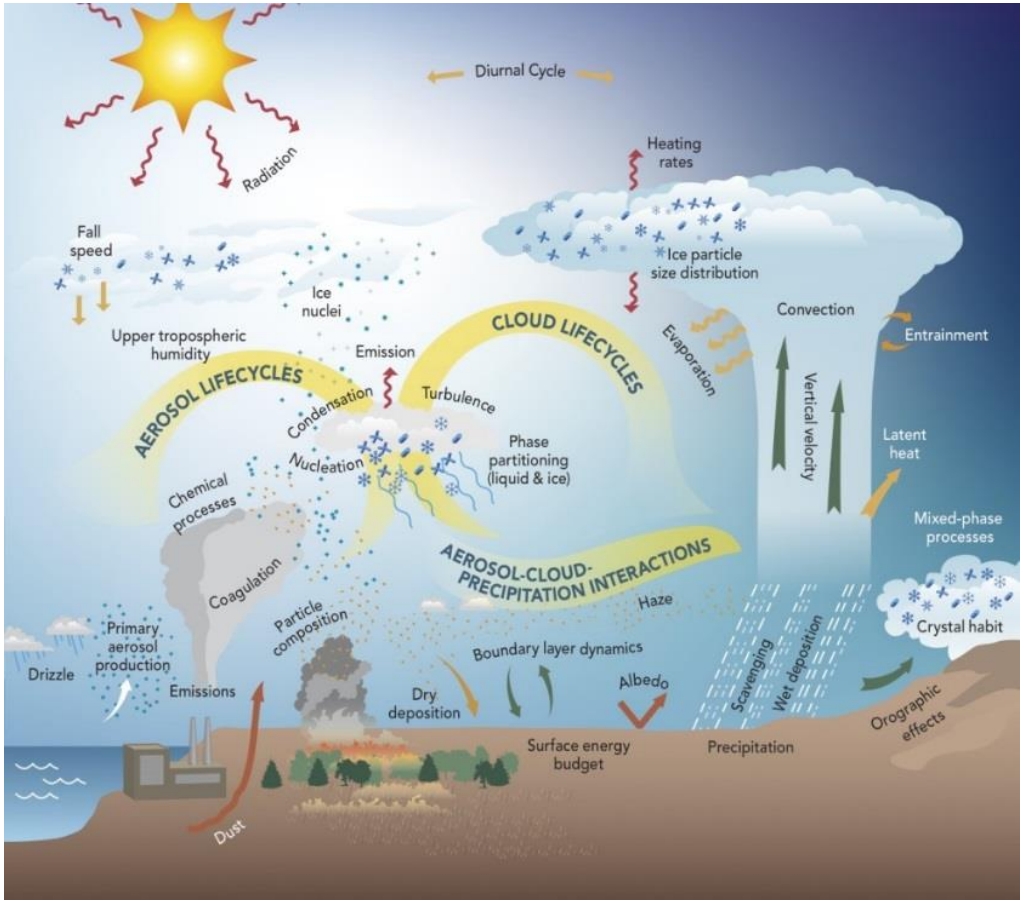
NILU

AEROCOM-PIII experiment: Aerosol lifetimes from Fukushima tracers

11 March 2011

Atmospheric lifetimes

The atmospheric lifetime of aerosols is **poorly constrained by observations** and there is a **large spread** in previous estimates and in models.

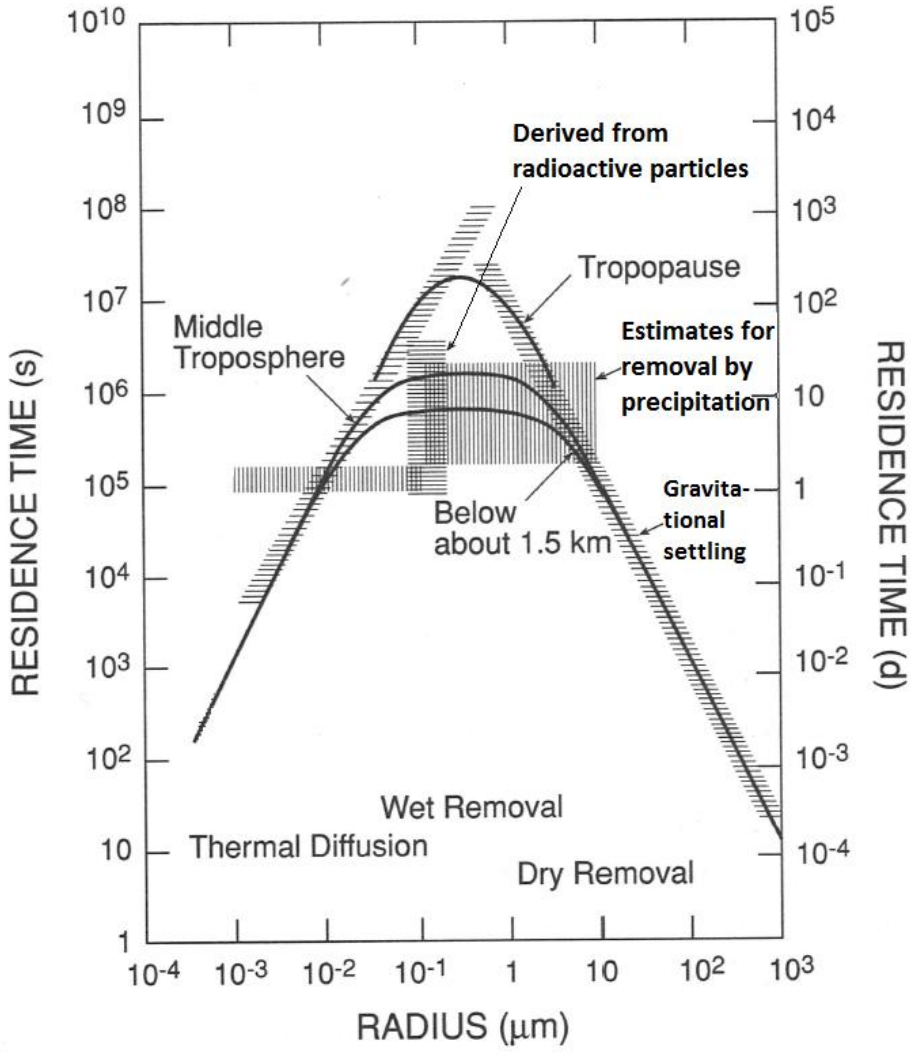


Credit: Atmospheric System Research (ASR) Science and Program Plan, 2010

Atmospheric lifetimes

Estimates for aerosol lifetimes have previously been derived from radioactive particles.

In this study we use radionuclides released from an emission pulse at the surface.

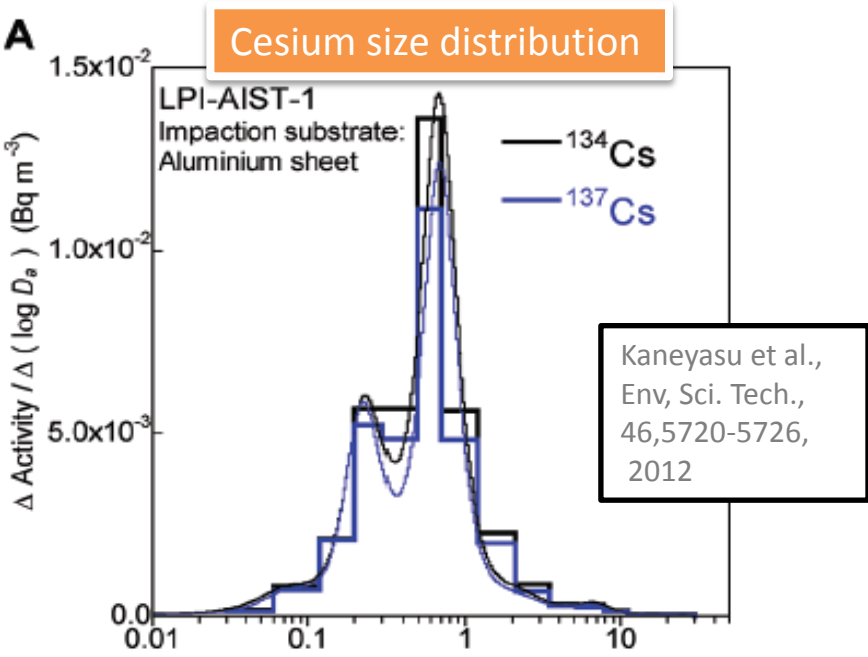


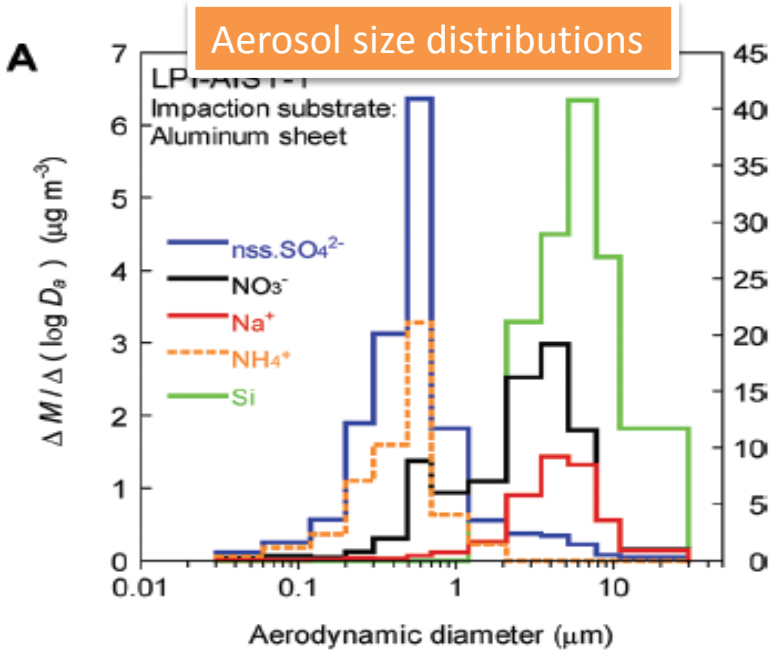
Modified from Jaenicke, R. (1988), Aerosol physics and chemistry.

"Tracers of opportunity"

Fukushima emissions

Cesium:
The radioactive isotope **cesium attaches to the ambient accumulation-mode aerosols**, particularly **sulphate** - and traces their fate in the atmosphere.





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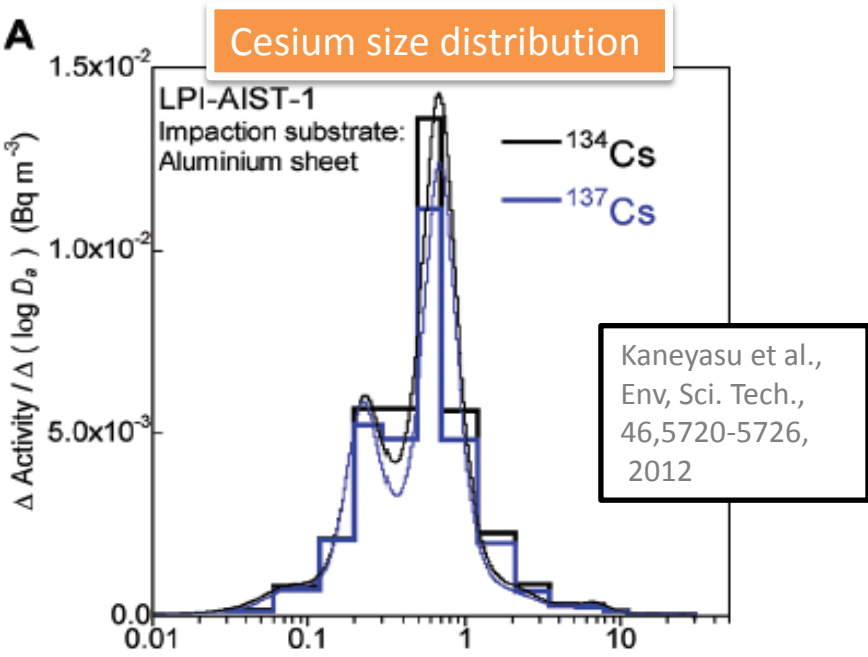
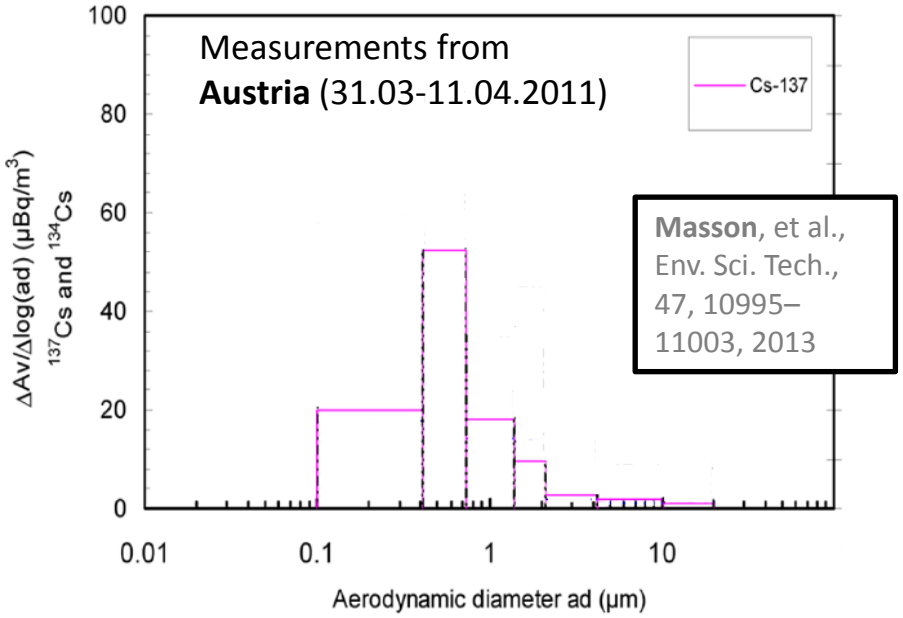
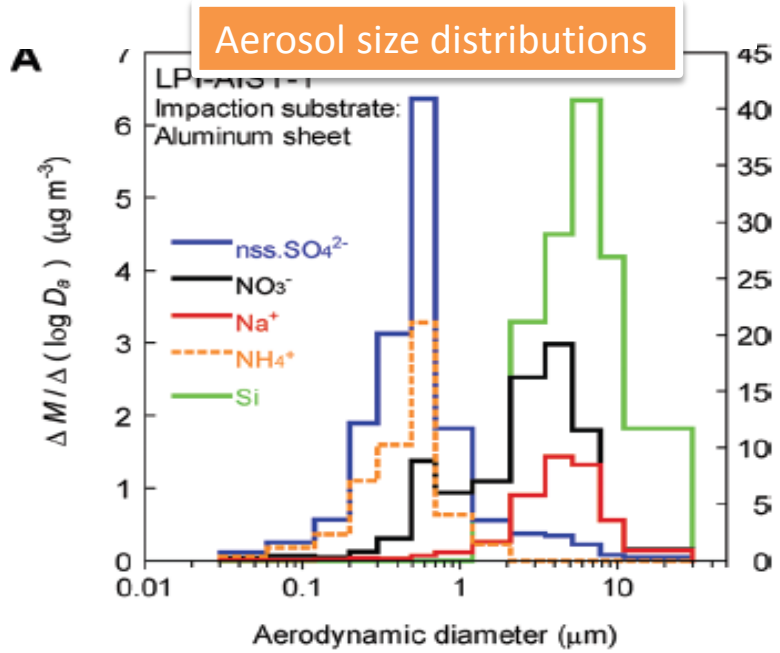




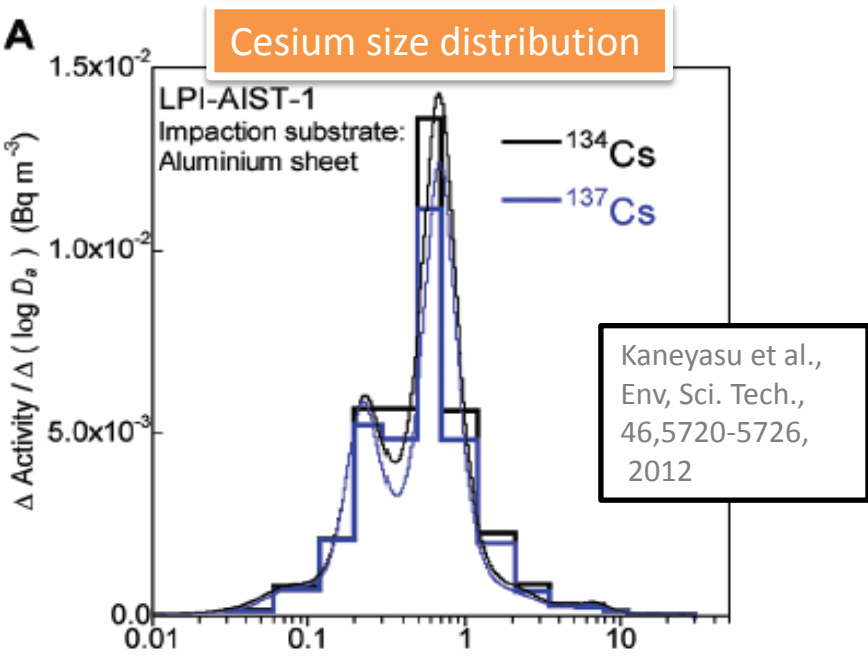


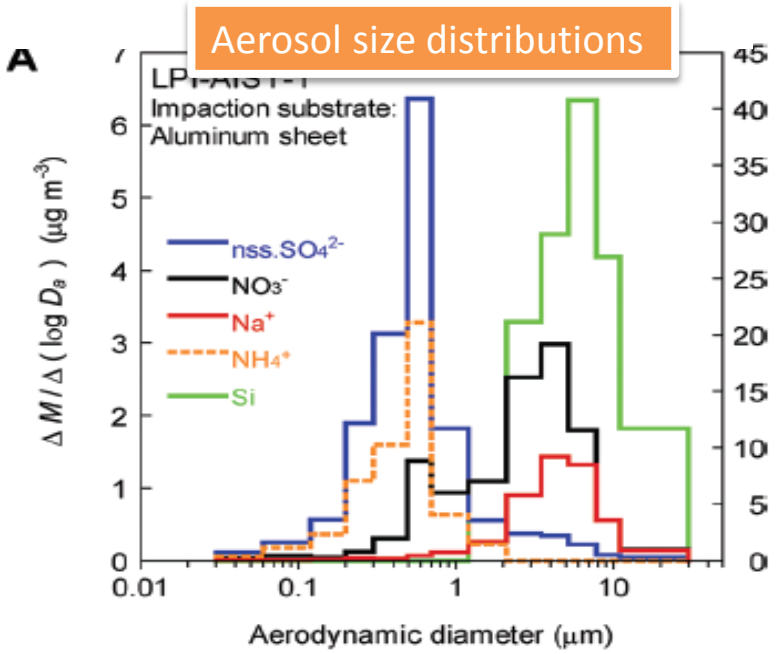
"Tracers of opportunity"

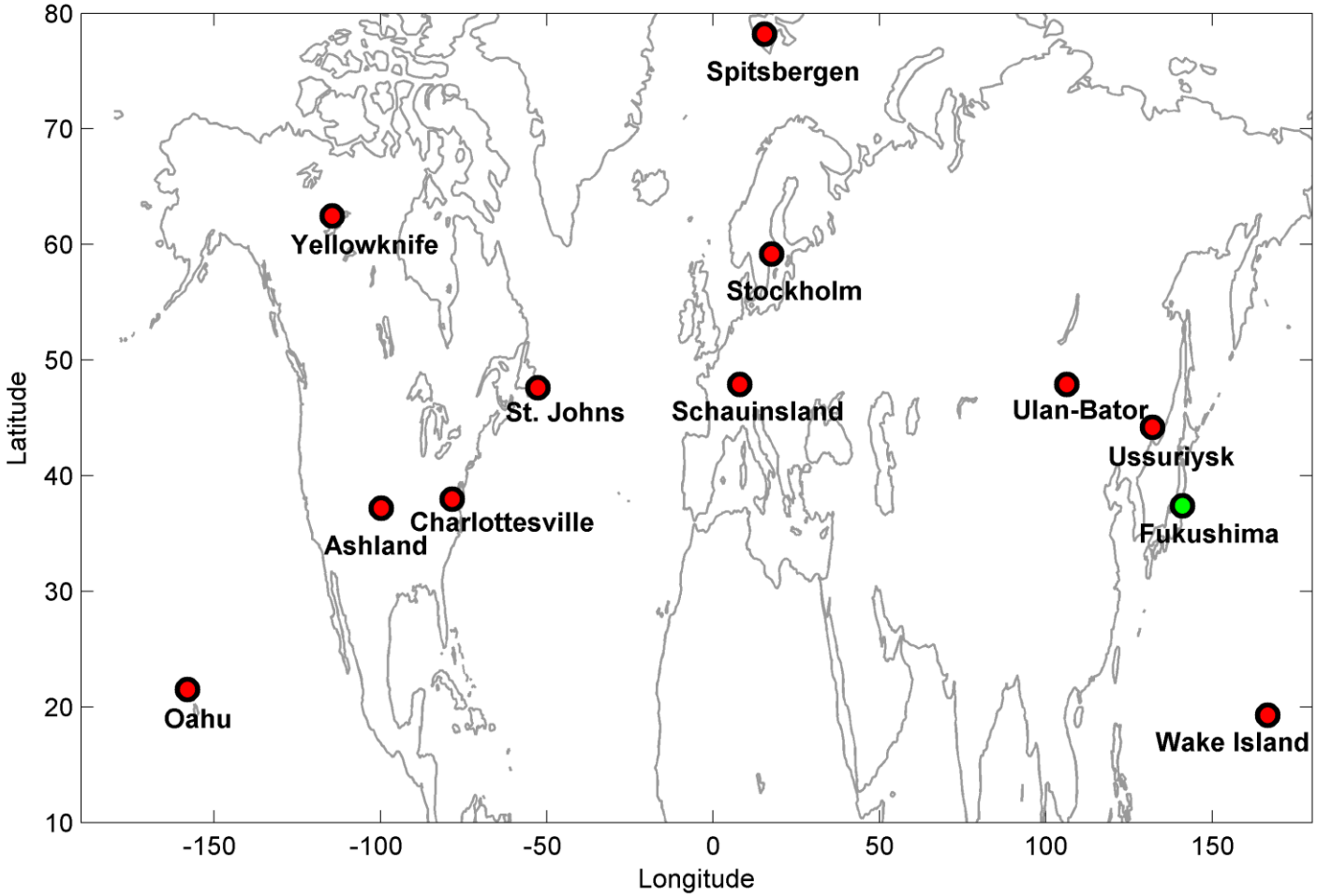
Fukushima emissions

Cesium:
The radioactive isotope **cesium attaches to the ambient accumulation-mode aerosols**, particularly **sulphate** - and traces their fate in the atmosphere.

Xenon:
Noble gas, tracer for atmospheric transport.



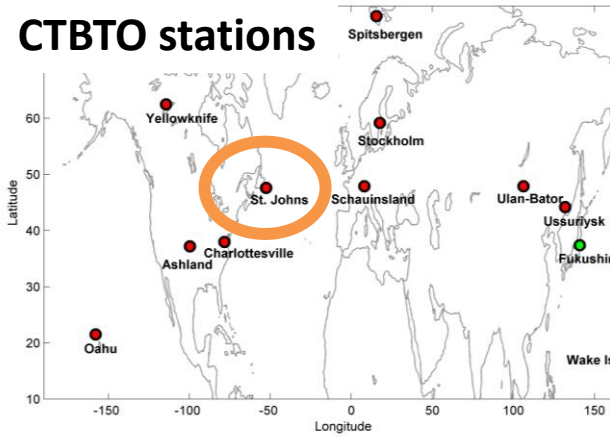


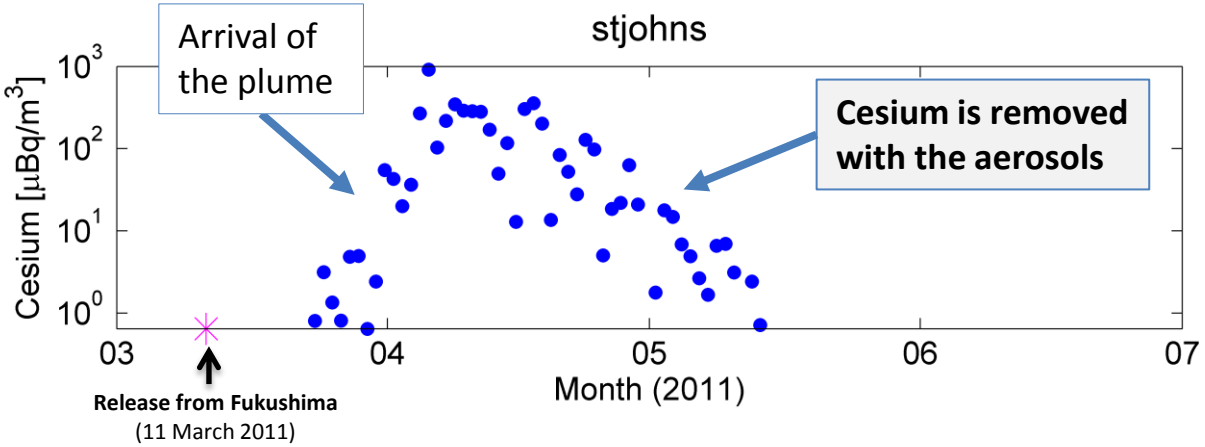


Comprehensive Nuclear-Test-Ban Treaty Organization (**CTBTO**) stations measuring radionuclides released from Fukushima

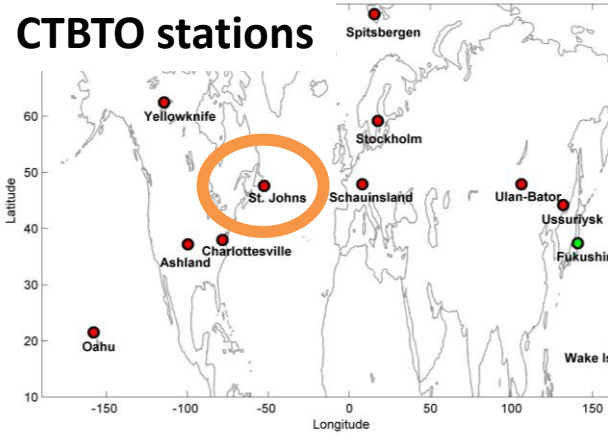
- over a period of 3 months!

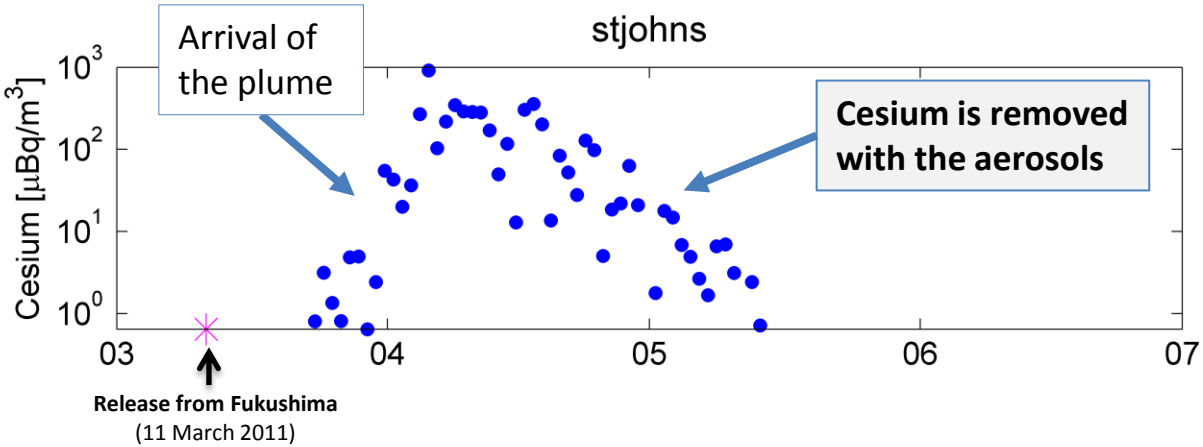
CTBTO stations

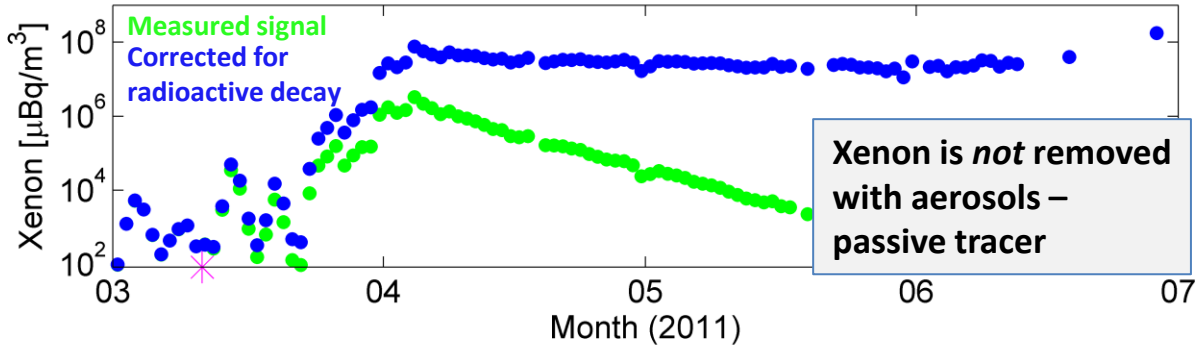




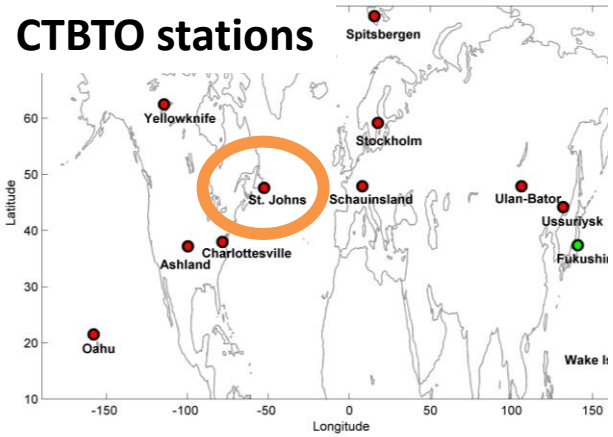
CTBTO stations

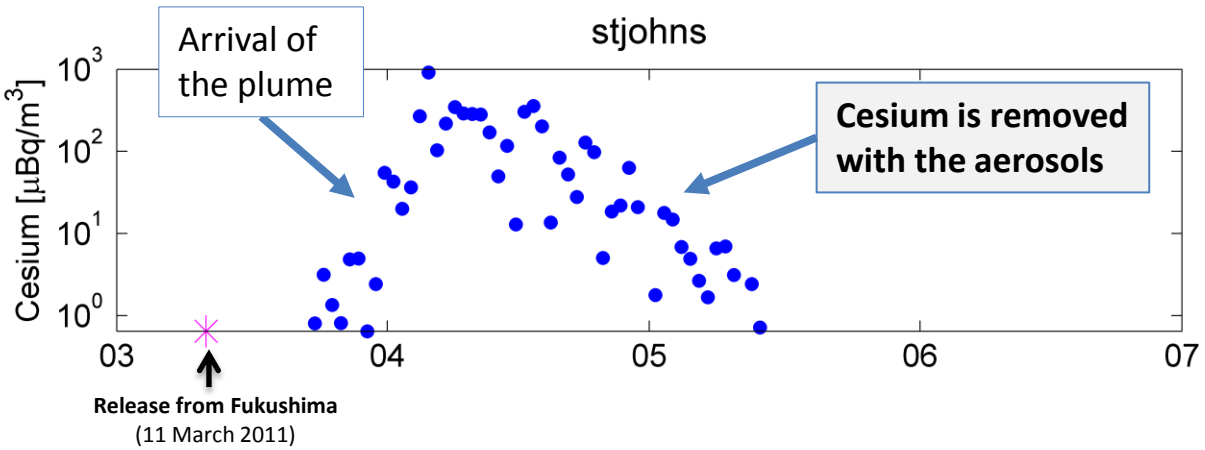


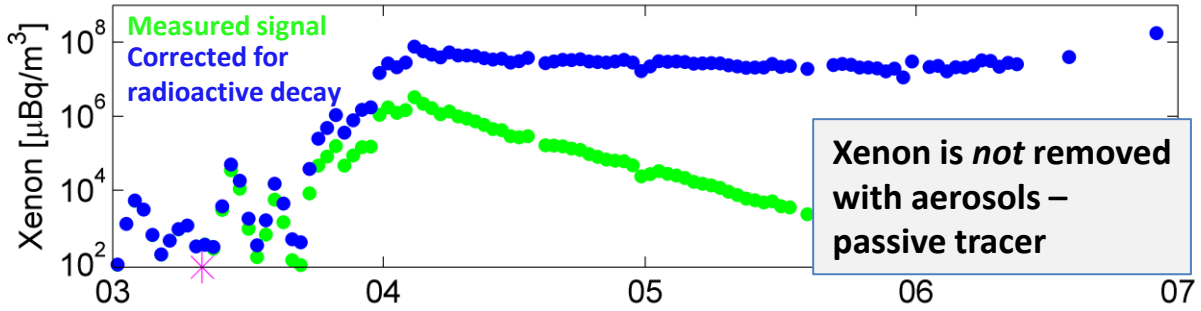


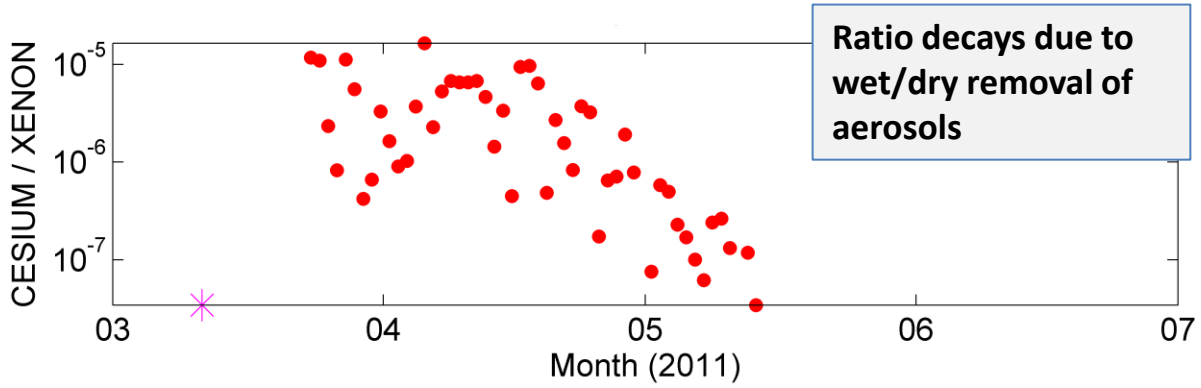


CTBTO stations





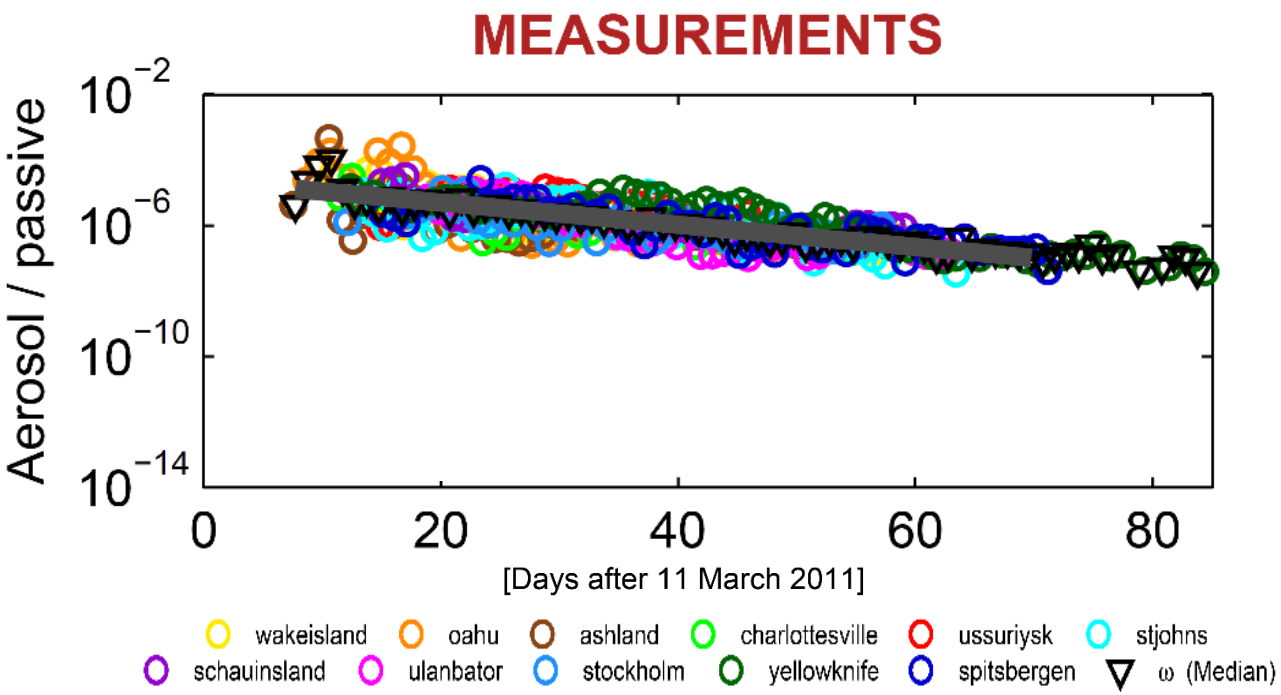


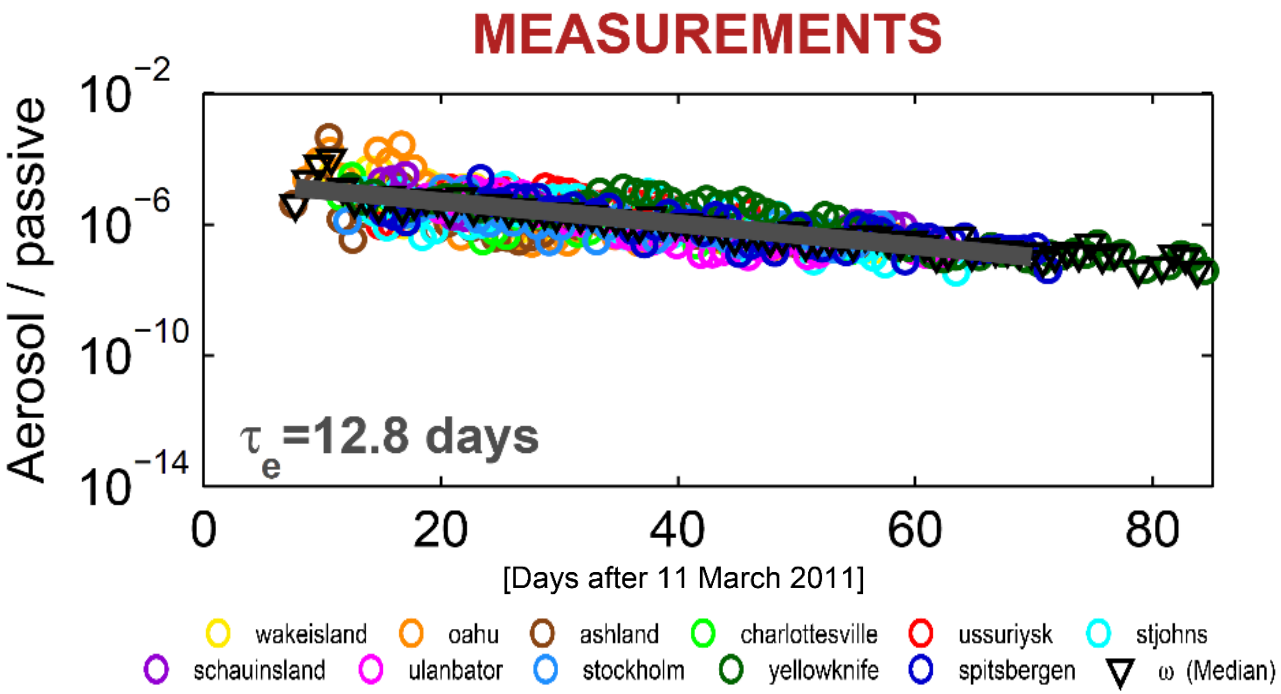


Ratio
Cesium/Xenon
Aerosol/Passive

Compensate for variations due to transport.

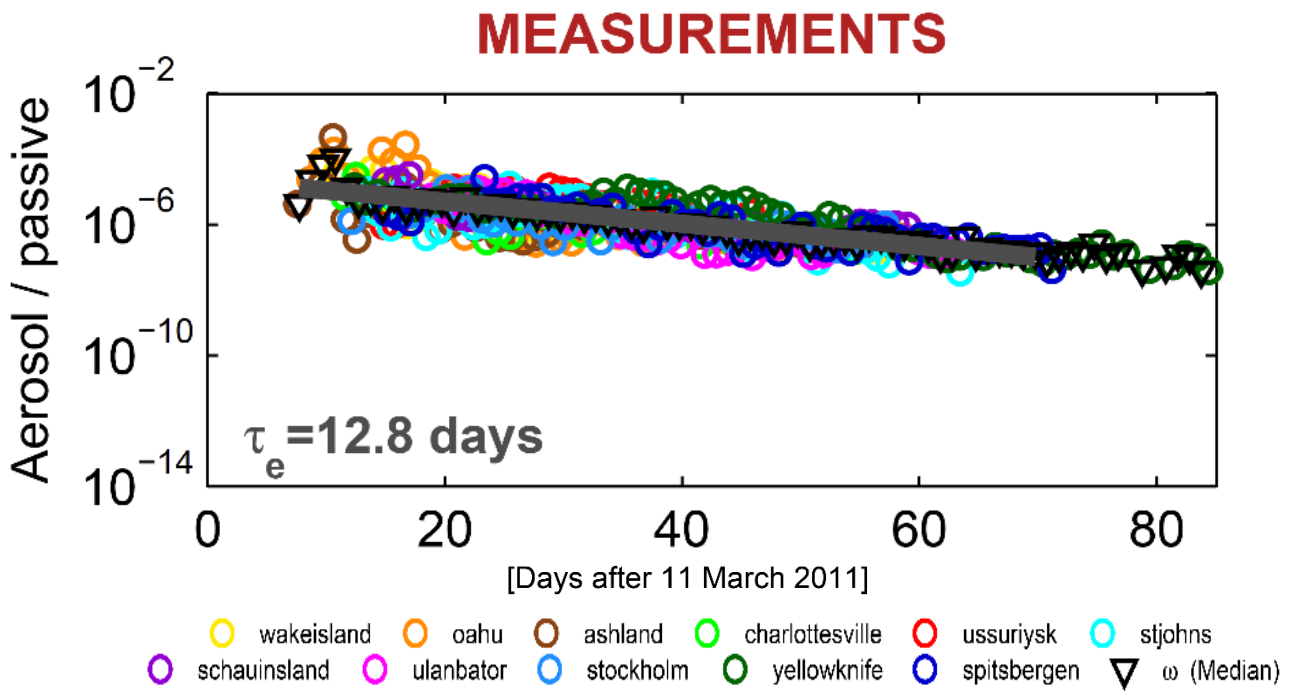
Loss of aerosols due to wet and dry removal only.


MEASUREMENTS
Aerosol / passive
10^{-2}
10^{-6}
10^{-10}
10^{-14}
0
20
40
60
80
[Days after 11 March 2011]
wakeisland
oahu
ashland
charlottesville
ussuriysk
stjohns
schauinsland
ulanbator
stockholm
yellowknife
spitsbergen
ω (Median)



- Fit of an exponential decay model through the ratios (cesium/xenon).
- **AM aerosol e-folding lifetime: 12.8 days**

Kristiansen, N. I., A. Stohl, G. Wotawa (2012)
Atmos. Chem. Phys., 12, 10759-10769,



- Fit of an exponential decay model through the ratios (cesium/xenon).
- **AM aerosol e-folding lifetime: 12.8 days**

Key Question
To what extent can models reproduce the observed loss of aerosol mass with time (i.e. aerosol lifetimes)?

Kristiansen, N. I., A. Stohl, G. Wotawa (2012)
Atmos. Chem. Phys., 12, 10759-10769,

	Models
1	**NorESM**
2	**GISS-ModelE2-TOMAS***
3	**GISS-ModelE-MATRIX***
4	**ULAQ-CCM**
5	**BCC_AGCM_2.0.1_CAM***
6	**LMDZORINCA**
7	**CAM5***
8	**CAM5_PNNL***
9	**CAM5_dynamic**
10	Echam5-Messy-Atmospheric-Circulation-Model (**EMAC-1**)
11	Echam5-Messy-Atmospheric-Circulation-Model (**EMAC-2**)
12	**ECHAM5-HAM2***
13	**ECHAM5-SALSA v5.4***
14	**GOES-CHEM v09-01-03**
15	**EEMEP** v2533
16	**OsloCTM2***
17	**OsloCTM3**
18	**NAME III**
19	**FLEXPART v9**

Models

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3	**GISS-ModelE-MATRIX***
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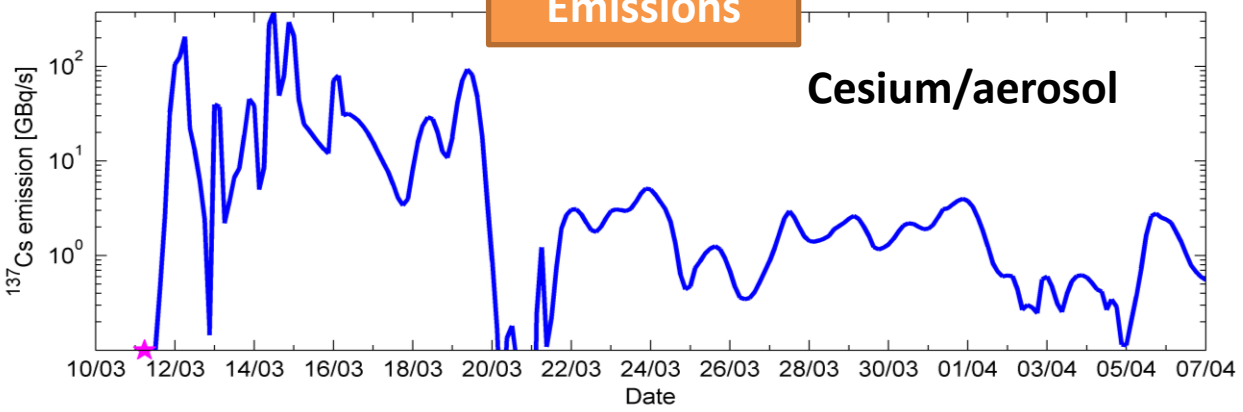
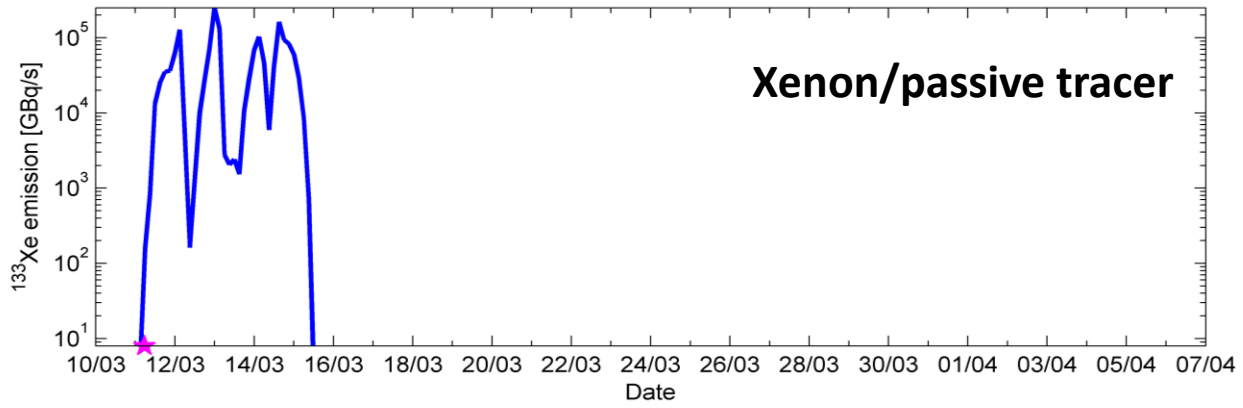
17	**OsloCTM3**
18	**NAME III**
19	**FLEXPART v9**

Emissions





Source terms estimated with inverse modeling
by Stohl et al., Atmos. Chem. Phys., 12, 2313-2343, 2012

	Models
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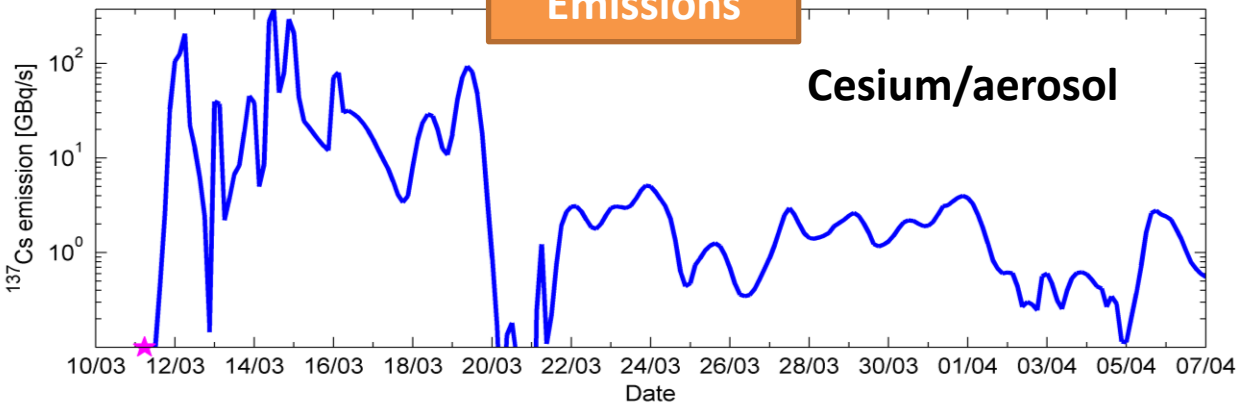
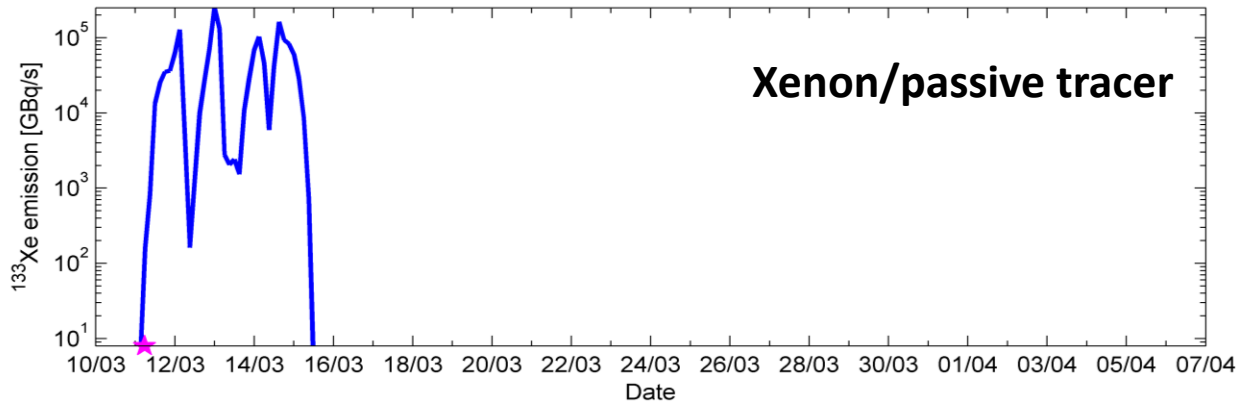
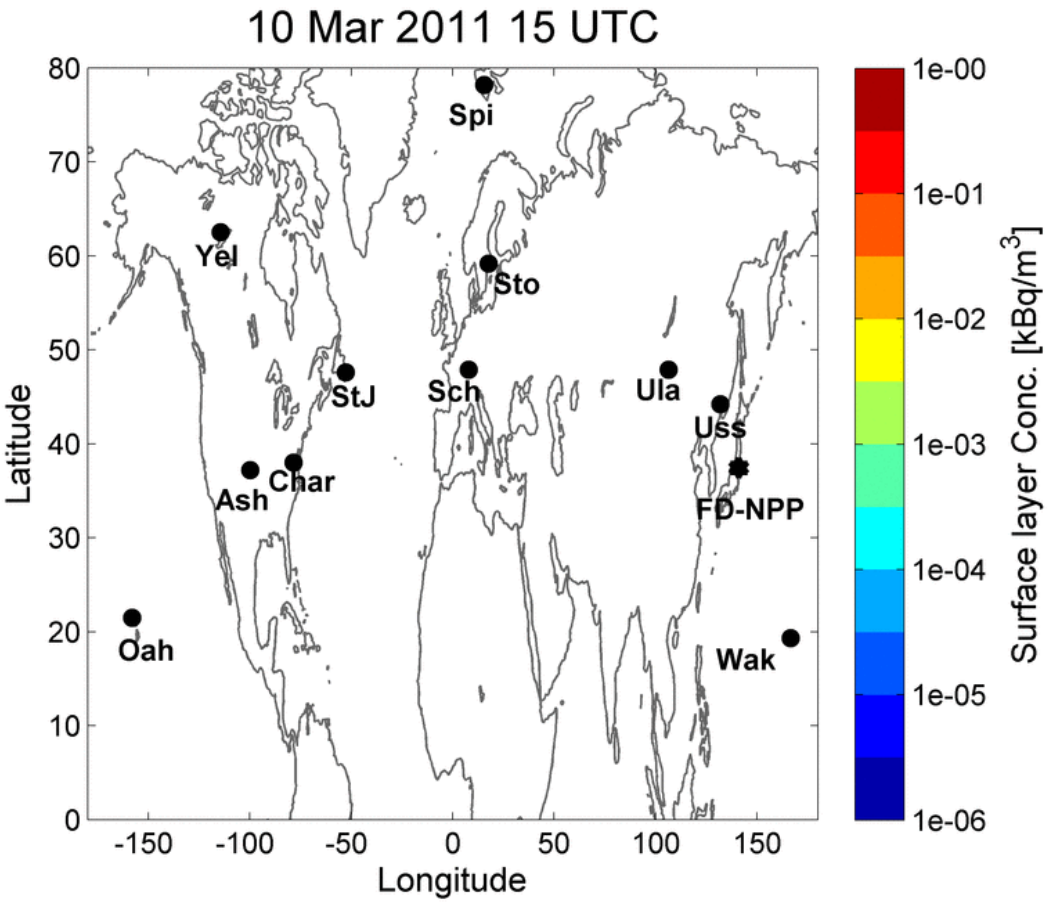
Emissions





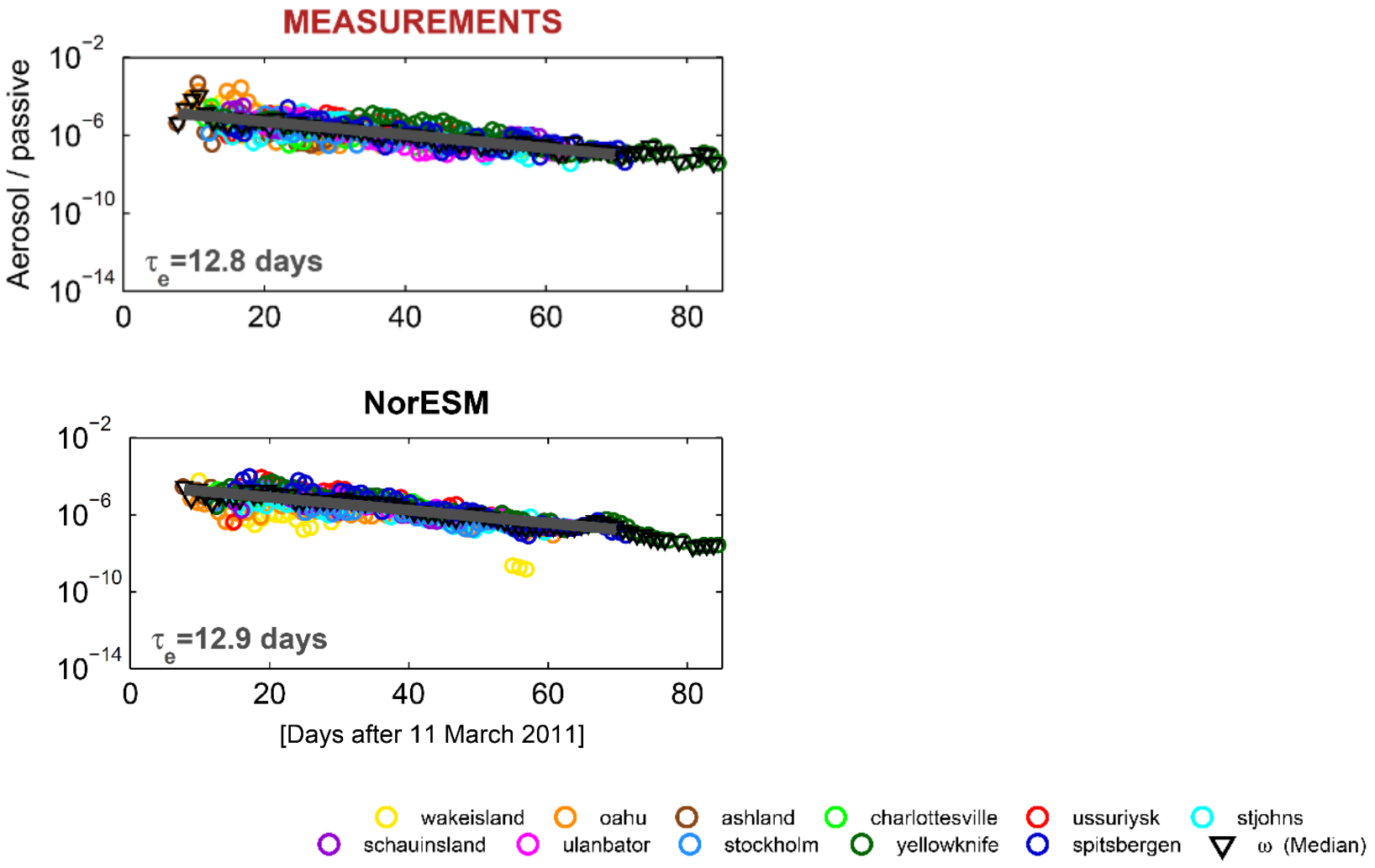
- The **cesium** was treated as **sulfate aerosols** in the model simulations, i.e. undergoing the same wet and dry deposition as accumulation-mode sulfate aerosols.
- The **xenon** was treated as a **passive tracer** without wet and dry removal processes.

- Model results were **sampled at exactly the same location and times as station measurements**
- This allows a direct comparison between measured and modelled aerosol decay.

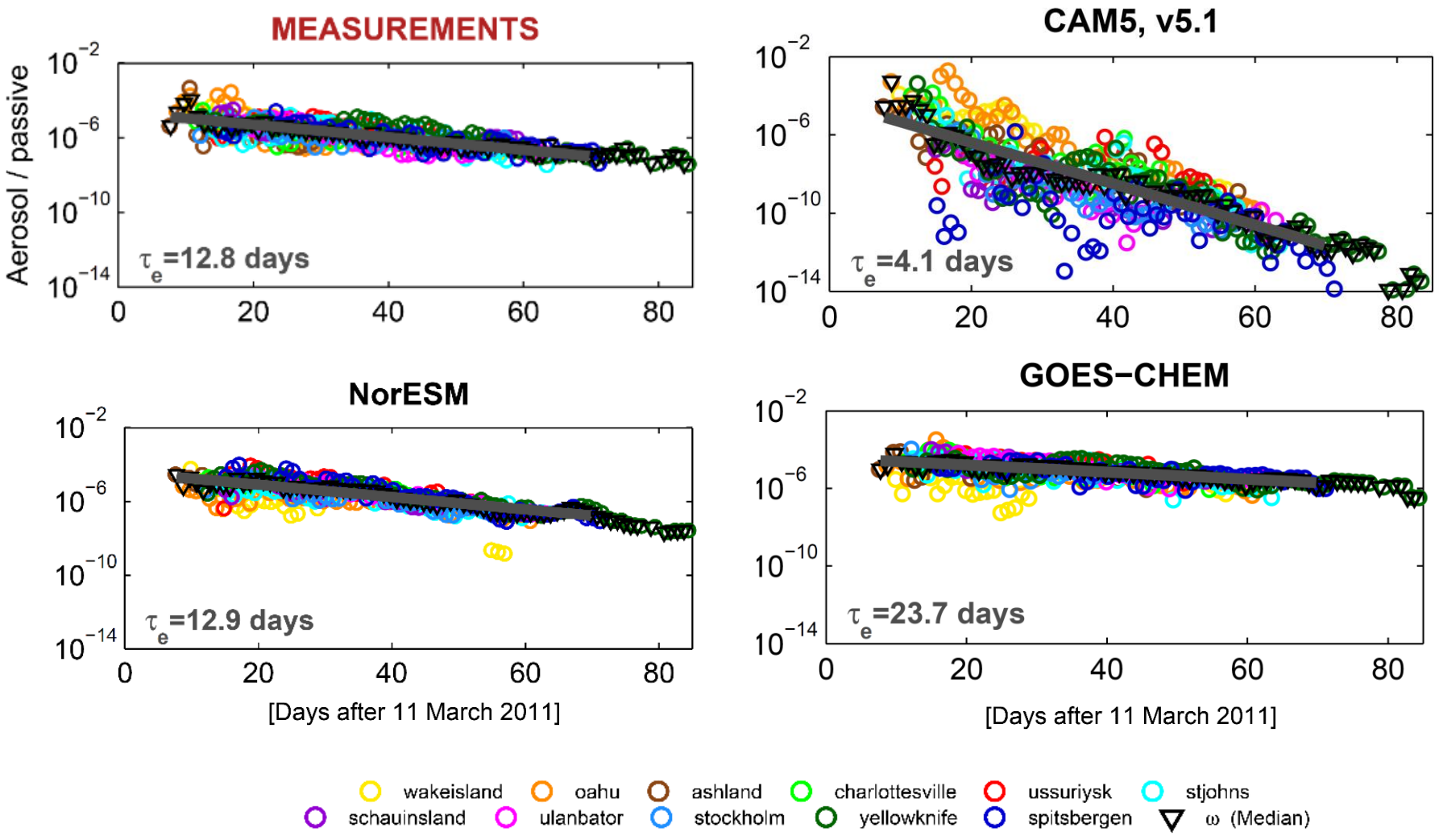


The transport of the radioactive cloud (xenon) across the Northern Hemisphere as simulated by the FLEXPART model using GFS meteorological data

Measured vs. modelled aerosol lifetimes



Measured vs. modelled aerosol lifetimes



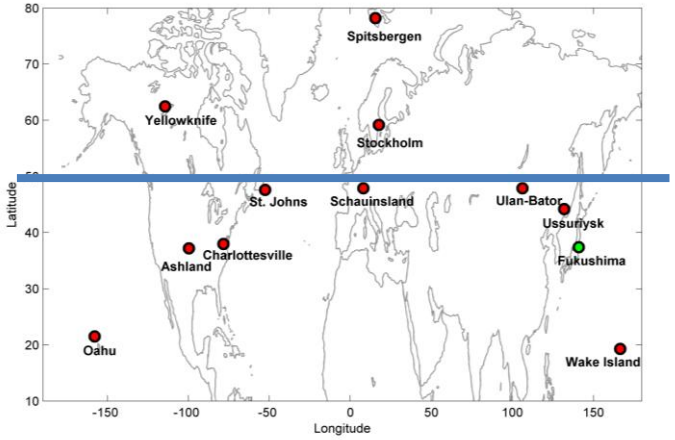
Measured vs. modelled aerosol lifetimes

1	NorESM	12.9
2	GISS-ModelE-TOMAS	10.3
3	GISS-modelE	11.2
4	ULAQ-CCM	5.6
5	BCC_AGCM	11.5
6	LMDZORINCA	12.9
7	CAM5	4.1
8	CAM5_PNNL	11.6
9	CAM5_dyn	7.7
10	EMAC-1	6.7
11	EMAC-2	7.5
12	ECHAM-HAM2	6.0
13	ECHAM5-SALSA	5.4
14	GOES-CHEM	23.7
15	EEMEP	17.0
16	OSLO-CTM2	10.0
17	OSLO-CTM3	8.3
18	NAME	8.9
19	FLEXPART	6.3
	MODEL MEDIAN ± STD	8.9±4.7
	OBSERVATIONS	12.8

- **Measured e-folding lifetime: 12.8 days** (95% confidence interval 11.7-14.1 days).
- The equivalent modelled lifetimes have a large spread, varying between 4.1 and 23.7 days with **a model median of 8.9±4.7 days.**
- Large variations in the modelled lifetimes are expected due to differences in the models' description of meteorology and especially tropospheric removal.

Latitudinal variation in aerosol lifetimes

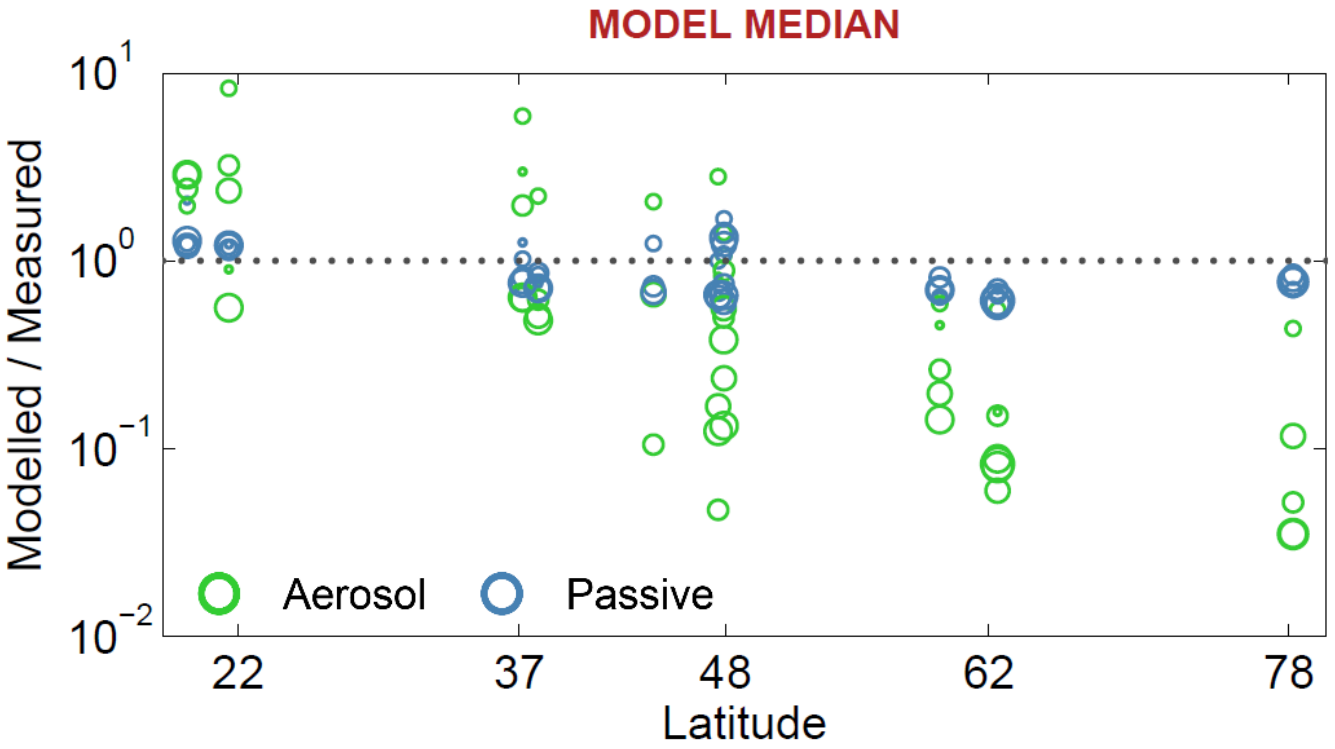
		Stations below 50°N	Stations above 50°N
1	NorESM	12.4	10.6
2	GISS-ModelE-TOMAS	9.5	10.4
3	GISS-modelE	9.7	12.0
4	ULAQ-CCM	5.9	5.2
5	BCC_AGCM	11.6	16.5
6	LMDZORINCA	11.9	14.1
7	CAM5	4.3	4.5
8	CAM5_PNNL	12.3	13.2
9	CAM5_dyn	7.3	10.3
10	EMAC-1	6.8	5.6
11	EMAC-2	6.8	6.7
12	ECHAM-HAM2	6.2	7.8
13	ECHAM5-SALSA	4.8	7.4
14	GOES-CHEM	19.4	29.0
15	EEMEP	14.7	31.0
16	OSLO-CTM2	9.4	19.8
17	OSLO-CTM3	8.5	8.8
18	NAME	9.0	9.8
19	FLEXPART	6.6	6.6
	MODEL MEDIAN ± STD	9.0±3.8	10.3±7.5
	OBSERVATIONS	11.5	14.7



- Both measured and median modelled lifetimes are **longer at higher latitudes,** suggests less efficient aerosol removal at high latitudes.

- Larger spread in model lifetimes and larger deviations from the measurements at high latitudes compared to at lower latitudes, **indicates larger uncertainties in the simulation of aerosol removal at higher latitudes.**

Model-obs deviations are largest for the aerosols at higher latitudes



- General **under-prediction** of both aerosol and passive tracer concentrations.
- Transport to the high latitudes is not strong enough in the models.
- Largest deviations for aerosol values suggest **aerosol scavenging is the major cause for disagreements with observations.**

Summary
AEROCOM-PIII Aerosol lifetime experiment

Aim
Evaluate measured and modelled accumulation-mode aerosol lifetimes.

Measurements
CTBTO station data of radioactive isotopes (aerosol-bound cesium, passive tracer xenon) released during the Fukushima accident of March 2011.

Models
19 global models simulated the transport of the radioactive isotopes using identical emissions.

Key question
To what extent can the models reproduce the observed loss of aerosol mass with time.

Results
- Measured e-folding lifetime**: 12.8 days** (11.7-14.1 days).
- The equivalent modelled lifetimes have a large spread, varying between 4.1 and 23.7 days with a model median of **8.9±4.7 days**.
- Models generally **under-predict** both aerosol and passive tracer concentrations, particularly at high latitudes. Largest under-predictions for aerosol values suggest aerosol scavenging is the major cause for disagreements with observations.