Evaluation of observed and modelled aerosol lifetimes

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

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Introduction

Aerosols have important impacts on air quality and climate, but the processes affecting their removal from the atmosphere are not fully understood and poorly constrained by observations. This makes modelled aerosol lifetimes uncertain. In this exercise, we address this uncertainty and try to get a better understanding of the causes of model differences.

Observations

Measurements of radioactive isotopes released during the Fukushima Dai-Ichi nuclear power plant accident of March 2011 are used as tracers of opportunity. The radioactive isotopes cesium (^{137}Cs) and xenon (^{133}Xe) were released in large quantities during the accident.

- **Cesium** attaches to ambient accumulation mode aerosols (especially sulfate) and traces their fate in the atmosphere.
- **Xenon** is a noble gas and behaves almost like a passive tracer and can serve as a reference species for atmospheric transport.

Atmospheric surface activity concentrations of cesium and xenon measured at 11 stations (Figure 1) operated by the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO), allow deriving a lifetime of the carrier aerosol (Kristiansen et al., 2012) which can be compared to aerosol lifetimes obtained by models (this study).

Model Simulations

For this study, 19 atmospheric transport models simulated the transport of the radioactive isotopes using *identical emissions*. The major emissions of the radionuclides occurred over about five days (11-15 March 2011) (Stohl et al., 2012). Some emissions of cesium continued until 20 March after which the emissions dropped significantly. 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 which is supported by studies showing that cesium mainly attaches to these aerosols (Kaneyasu et al., 2012; Masson et al., 2013) . The xenon was treated as a passive tracer without wet and dry removal processes. Model results sampled at exactly the same location and times as station measurements allow a direct comparison between measured and modelled aerosol decay and provide a strong constraint on the modelled aerosol lifetime.

Key Question

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

Main Results

- Figure 2 and Table 1: From station measurement data the **measured e-folding lifetime τ_e is 12.8 days** (95% confidence interval 11.7-14.1 days). The equivalent modelled τ_e lifetimes have a large spread, varying between 4.1 and 23.7 days with **a model median of 8.9±4.7 days**.
- Instantaneous lifetimes show that the initial removal was quicker due to the emissions occurring at low altitudes and co-location of the plume with strong precipitation.
- Deviations between measured and modelled aerosol lifetimes are **largest for the northernmost stations** and at later time periods.
- Comparisons of measured and modelled concentrations at each measurement station show a **general underestimation of both aerosol and passive transport tracer concentrations**. Transport to the high latitudes is not strong enough in the models, but the aerosol underestimation is larger indicating that problems with the aerosol scavenging is the major reason for disagreements with observations.

Measurement network and transport of the radioactive cloud from Fukushima

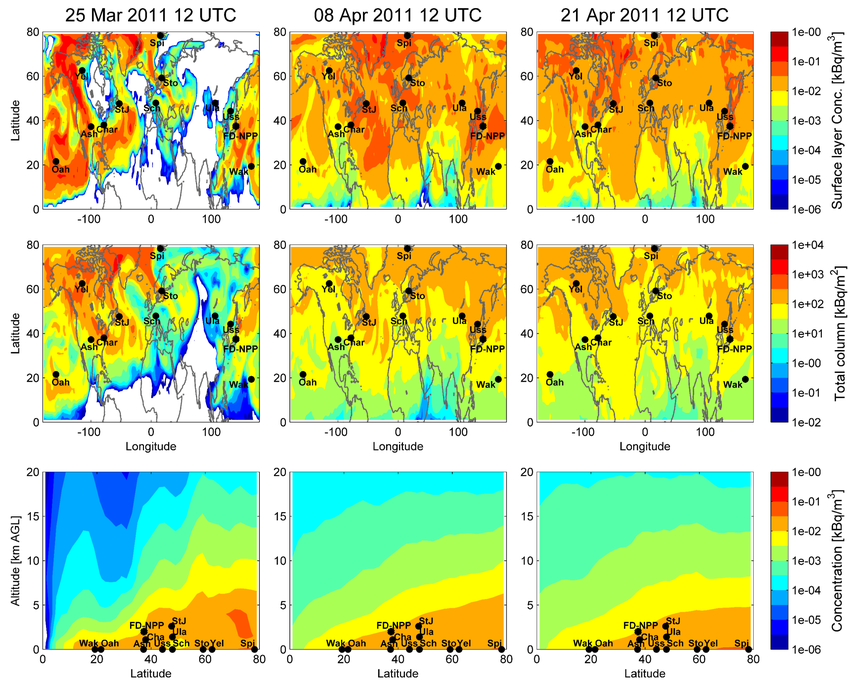

Figure 1: *The transport of the radioactive cloud across the Northern Hemisphere as simulated by the FLEXPART model using GFS meteorological data. ^{133}Xe surface concentrations (upper panel), total atmospheric columns (middle panel) and zonal mean vertical distribution (lower panel) for 25 March, 8 April and 22 April 2011 (2, 4 and 6 weeks, respectively, after the start of the initial release on 11 March). The 11 CTBTO stations are marked with black circles and the position of the Fukushima Dai-Ichi Nuclear Power Plant (FD-NPP) with a black star.*

Direct comparison of measured and modelled aerosol decay

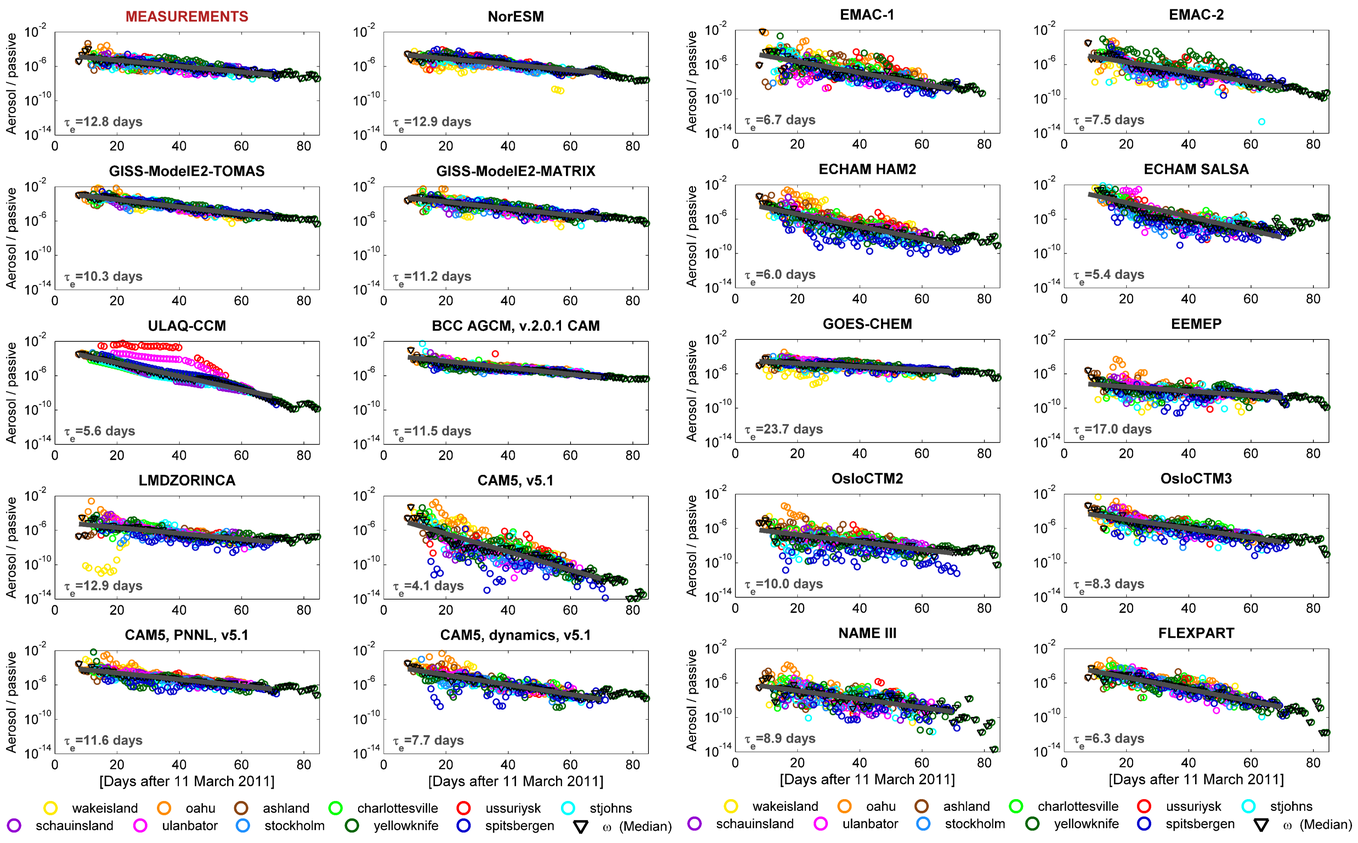

Figure 2: *Measured and modelled ratios between aerosol-bound cesium (^{137}Cs) and passive tracer xenon (^{133}Xe), with time at the 11 CTBTO station locations. ω (Median, black triangles) represent the daily median ratios (median of activity concentrations for each day over all stations). Fits of exponential decay models to the ω data are shown as grey lines with e-folding time scales τ_e as indicated.*

	MODEL	e-folding lifetime τ_e [days]				
		All days All stations	All days Stations below 50°N	All days Stations above 50°N	Day 25-45 All stations	Day 45-70 All stations
1	NorESM	12.9	12.4	10.6	9.9	19.1
2	GISS-ModelE-TOMAS	10.3	9.5	10.4	9.3	14.3
3	GISS-modelE	11.2	9.7	12.0	9.1	18.6
4	ULAQ-CCM	5.6	5.9	5.2	5.8	3.8
5	BCC_AGCM	11.5	11.6	16.5	17.3	11.9
6	LMDZORINCA	12.9	11.9	14.1	7.1	23.4
7	CAM5	4.1	4.3	4.5	11.1	3.3
8	CAM5_PNNL	11.6	12.3	13.2	14.2	12.4
9	CAM5_dyn	7.7	7.3	10.3	6.5	8.2
10	EMAC-1	6.7	6.8	5.6	9.4	6.4
11	EMAC-2	7.5	6.8	6.7	10.3	6.7
12	ECHAM-HAM2	6.0	6.2	7.8	6.2	6.3
13	ECHAM5-SALSA	5.4	4.8	7.4	9.4	34.7
14	GOES-CHEM	23.7	19.4	29.0	24.2	24.9
15	EEMEP	17.0	14.7	31.0	19.4	21.6
16	OSLO-CTM2	10.0	9.4	19.8	13.7	11.3
17	OSLO-CTM3	8.3	8.5	8.8	12.2	8.4
18	NAME	8.9	9.0	9.8	7.3	5.5
19	FLEXPART	6.3	6.6	6.6	6.7	5.6
	MODEL MEDIAN ± STD	8.9±4.7	9.0±3.8	10.3±7.5	9.4±4.9	11.3±8.6
	OBSERVATIONS	12.8	11.5	14.7	12.4	18.5

Table 1*. E-folding aerosol lifetimes estimated from the decay of the aerosol/passive ratios at 11 station locations. Variations in lifetime for different latitude regions (below and above 50°N) and time periods are also given (grey values). The data are shown in Figure 2.*

Aerosol to passive tracer ratio

To evaluate the loss of aerosols due to wet and dry removal only, we normalize the cesium values by the xenon values.
This largely compensates for variability in transport. Hence, the ratio of the aerosol-bound cesium to the passive tracer xenon is used throughout the lifetime evaluations.

E-folding lifetime

The e-folding lifetime is used as means of evaluation:

$$\tau_e = \frac{-t_i}{ln\frac{C(t_i)}{C(t_0)}}$$

$C(t_i)$ is the ratio at time t_i, $C(t_0)$ the initial ratio at time t_0, and t_i is the time since t_0.

E-folding lifetime is *not* directly comparable to global mean aerosol lifetimes (Croft et al., 2014).

Sensitivity Studies, Open Questions and Further Work

- The model-observation deviations can be due to errors in
  - convective transport and scavenging
  - mixing from free troposphere to the boundary layer
  - the assumption that cesium attached only to sulfate and neglecting coagulation with larger aerosols.

- Five ensemble members of the NorESM model show no large differences in lifetime, suggesting that the lifetime estimates are independent of the meteorological situation. Also, as shown by Croft et al. (2014), the e-folding lifetimes do not depend very much on the exact model setup (emission altitude, location and time). Therefore, the model's scavenging parameterizations are likely the main cause of model differences.

- How sensitive are the results to the sampling at the station locations and measurement times (e.g. Croft et al. 2014)?

- Over which days to do the optimal fit of the exponential decay models? First few days only a few stations sampled the radionuclides, later on the detection limit is a problem.

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