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Atmospheric lifetime of caesium-137 as an estimate of aerosol lifetime –quantified from global measurements in the months after the Fukushima Dai-ichi nuclear accident

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Radionuclides like caesium-137 (137Cs) can be emitted to the atmosphere in great quantities during nuclear accidents and are of significant health impact. A global set of radionuclide measurements collected over several months after the accidental release from the Fukushima Dai-ichi nuclear power plant in March 2011 has been used to estimate the atmospheric lifetime of 137Cs. Lifetime is here defined as the e-folding time scale (the time interval in which the exponential decay of the 137Cs quantity has decreased by factor of e).

The estimated atmospheric lifetime of 137Cs can also be used as an estimate of the lifetime of aerosols in the atmosphere. This is based on the fact that 137Cs attaches to the ambient accumulation-mode (AM) aerosols and trace their fate in the atmosphere. The 137Cs "tags" the AM aerosols and both the 137Cs and AM aerosols are removed simultaneously from the atmosphere by scavenging within clouds, precipitation and dry deposition. The 137Cs emitted from Fukushima attached mainly to sulphate aerosols in the size range 0.1–2 μm diameter.

Measured 137Cs activity concentrations from several stations spread mostly over the Northern Hemisphere were evaluated, and the decrease in activity concentrations over time (after correction for radioactive decay) reflects the removal of aerosols by wet and dry deposition. Corrections for air mass transport were made using measurements of the noble gas xenon-133 (133Xe) which was also released during the accident. This noble gas does not attach to the aerosols and was thus used as a passive tracer of air mass transport.

The atmospheric lifetime of 137Cs was estimated to 10.0-13.9 days during April and May 2011. This represents the atmospheric lifetime of a "background" AM aerosol well mixed in the extratropical northern hemisphere troposphere. It is expected that the lifetime of this vertically mixed background aerosol is longer than the lifetime of fresh AM aerosols directly emitted from surface sources. Possible caveats like late emissions and resuspension were found not to significantly affect the results.

The estimated lifetimes from this study are within the much larger and uncertain range of previously observation-based studies of aerosol lifetimes (less than 4 days to more than a month). However, modelled aerosol lifetimes from air quality and climate models typically range 3-7 days which is substantially lower than the mean AM lifetimes obtained from this study. The difference points towards a too quick removal of AM aerosol in the models and further research on the cause of this discrepancy is warranted. Too short modelled AM aerosol lifetimes would have serious implications for air quality and climate model predictions. By running several major climate and air quality models for the Fukushima case, an evaluation of the models performance compared to the measurements can be directly obtained.