Interaction of Meteoric Smoke Particles with stratospheric H_2SO_4 and spatial variations in surface deposition

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Abstract

There are large uncertainties in the transport and surface deposition of upper atmospheric particles used to construct climate proxies. Two such examples are meteoric smoke particles (MSPs) and Beryllium-10. We have used a 3D CCM to simulate the transport and deposition of Plutonium-238 oxide nanoparticles formed after the ablation of a power unit in the upper stratosphere (~11ºS) in 1964. The model reproduces both the observed hemispheric asymmetry and timescale of Pu-238 deposition. We then use the CCM to investigate the transport of MSPs from the upper mesosphere. The strongest MSP deposition is predicted to occur at mid-latitudes, providing a significant source of Fe fertilization to the Southern Ocean. The model also predicts more deposition in Greenland than Antarctica (by a factor of ~15, in agreement with ice core measurements), showing that climate proxy measurements from a limited number of sites must be interpreted with care.

1. Background

MSPs are nm-sized particles that form in the upper mesosphere from the condensation of vapour produced by meteoric ablation. Our aim is to understand MSP transport and surface deposition, in order to interpret the recent measurements of extra-terrestrial elements, including Ir, Pt and super-paramagnetic Fe, that have accumulated in polar ice cores. Measurements in ice cores in central Greenland, Vostok and EPICA-Dome C show that the deposition rate in Greenland is ~15 times higher than at Vostok and EPICA. The Greenland estimate of the total input of interplanetary dust particles into the earth's atmosphere is around 200 t d^{-1} (Gabrielli *et al*., 2004). This is significantly higher than most estimates based on observations within the atmosphere (>70 t d^{-1} , Plane *et al*., 2012).

We use surface deposition of Pu-238 particles which were injected in the stratosphere on April 21st, 1964, when a US Transit navigational satellite launched failed to reach orbital velocity. The payload included a SNAP-9A radioisotope thermoelectric generator, containing 17 kilocuries (~1 kg) of Pu-238 ($t_{½}$ = 88 years), which reentered the atmosphere around 11º S over the Indian Ocean. Due to the uniqueness of the SNAP Pu-238 isotope the spatial surface distribution of its surface deposition could be established from soil data at 65 sites.

2. Model Setup

Run	Input location/ type	Altitude
A_35km12S	SNAP @12S, one off	35km
B_35km15S	SNAP @15S, one off	35km
C_35km17S	SNAP @17S, one off	35km
D_45km15S	SNAP @15S, one off	45km
E_55km15S	SNAP @15S, one off	55km
F_MSP	MSP, global, continuous	80km

Table 1: Different initial conditions used in UMSLIMCAT to study transport and surface deposition of the SNAP particles and MSPs.

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3. Zonal mean distribution after one month

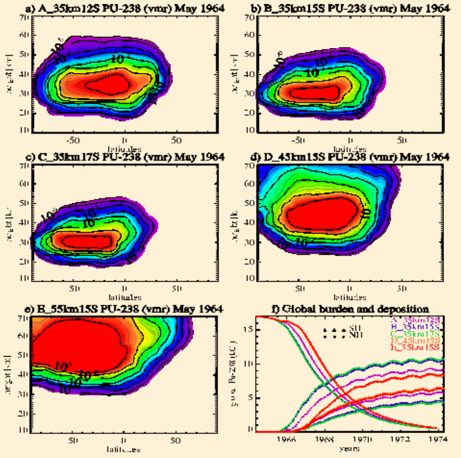

Fig 1. Modelled zonal mean mixing ratios of Pu-238 particles (×10^{12} mol mol^{-1}) at the end of May 1964 for five model runs. Lower right panel: time series of global atmospheric burden of Pu-238 (solid lines in kCi) and deposited Pu-238 (in kCi) in SH (triangles) and NH (open circles) from five model runs.

4. Surface Deposition of Pu-238 particles

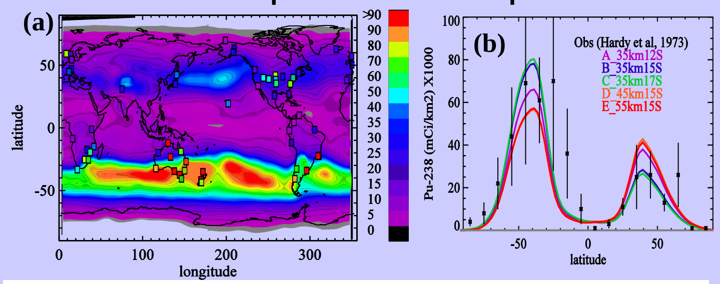

Fig 2. (a) Modelled deposition map (mCi km^{-2}) from run B_35km15S. Measurement sites are indicated by the coloured boxes. (b) Comparison of the zonal mean modelled deposition (mCi km^{-2}) from the 5 model runs with observations (Tables 1 and 2 from Hardy *et al*., 1973*)*.

5. Surface Deposition of MSP particles

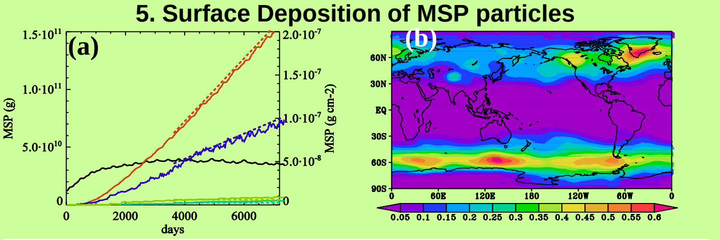

Fig 3. (a) Temporal evolution of total atmospheric mass burden (g, left hand axis) of MSPs (black line) and the accumulated surface-deposited MSP mass (orange line). The deposition fluxes (g cm^{-2}, right-hand axis) are shown for GRIP (blue line), VOSTOCK (turquoise line) and DOME-C (green line). (b) Map of annual mean Fe deposition rate (μmol Fe m^{-2} y^{-1}) from run F_MSP. Over the Southern Ocean, where the supply of bio-available iron to phytoplankton is limited. The estimated input into the Southern Ocean from the model is ~0.4 μmol Fe m^{-2} y^{-1}, and is comparable with an Aeolian dust input of ~30 μmol Fe m^{-2} y^{-1} as the MSP Fe should be in the form of highly soluble ferrous/ferric sulphate after processing in the stratospheric sulphate layer.

6. Summary and Conclusions

- **Surface deposition of Pu-238 particles is in agreement with the observations→ provides confidence in UMSLIMCAT transport and wet deposition scheme.**
- **Model suggests 10 times more deposition at GRIP than VOSTOCK→ consistent with ice core measurements of superparamagnetic Fe and Ir/Pt.**
- **But, a factor of 3–4 times larger meteoric ablation rate required to model the measured MSP flux, as opposed to the meteoric metal layers, and this is a discrepancy which needs to be resolved in order to better quantify the deposition of bioavailable cosmic Fe to the Southern Ocean.**

7. References

Dhomse et al (2013) **GRL** 40, 4454-4458. **Gabrielli** et al (2004) **Nature** 432, 1011-1014. **Hardy** et al (1973) **Nature** 241, 445-446. **Saunders** et al (2012) **ACP** 12, 4387-4398. **Plane** et al (2012) **Chem. Soc. Rev.** 41, 6507-6518.