

S1 Global emissions from source categories

Table S1: Compilation of literature estimates of global emission fluxes of selenium from different sources (Gg Se yr^{-1}). Minimum values for each source are highlighted in blue and maximum values are highlighted in red. No volcanic emission estimates are highlighted, since the range of volcanic Se emissions used in the sensitivity analysis is calculated from Table S2.

| Source | Marine | Terrestrial | Anthropogenic | Volcanic |
|---------------------------------------|---------|-------------|---------------|----------|
| Mosher and Duce (1987) | 5.0–8.0 | 0.7–3.6 | 6.0 | 0.4–1.2 |
| Nriagu and Pacyna (1988) ^a | | | 3.0–9.6 | |
| Nriagu (1989) | 0.4–9 | 0.15–5.25 | | 0.1–1.8 |
| Amouroux and Donard (1996) | 28.5 | | | |
| Amouroux et al. (2001) | 35 | | | |

^a Calculated for total Se emissions, since Nriagu and Pacyna (1988) only list particulate emissions range.

S2 Volcanic emissions

Table S2: Comparison of selenium to sulfur ratios in volcanic emissions, extended from Floor and Román-Ross (2012). The minimum Se:S ratio is highlighted in blue and maximum value is highlighted in red.

| Volcano | Period | Se:S ratio ($\times 10^4$) | Reference |
|-----------------------------|------------------|---------------------------------|------------------------------------|
| Mount Erebus, Antarctica | 1986–1991 | 0.46 | Zreda-Gostynska et al. (1997) |
| | 1997–2000 | 0.11 | Wardell et al. (2008) |
| White Island, New Zealand | 2000–2001 | 0.06 | Wardell et al. (2008) |
| Merapi, Indonesia | 1984 (Jan–Feb) | 39 | Symonds et al. (1987) |
| Kurile Island, Russia | 1990–1993 | 0.10 | Taran et al. (1995) |
| Kilauea, USA | 1983 (Nov) | 0.60 | Finnegan et al. (1989) |
| | 1983–1984 | 18 | Olmez et al. (1986) |
| | 1984–1996 | 0.64 | Hinkley et al. (1999) |
| St Helens, USA | 1980 (May) | 1.3 | Vossler et al. (1981) |
| | 1980 (September) | 3.6 | Phelan et al. (1982) |
| Augustine, USA | 1976 | 3.1 | Lepel et al. (1978) |
| Soufrière Hills, Montserrat | 1996 | 0.72 | Allen et al. (2000) |
| Stromboli, Italy | 1993–1997 | 0.84 | Allard et al. (2000) |
| Etna, Italy | 1976 | 15 | Faivre-Pierret and Le Guern (1983) |
| | 1976 (June) | 6.8 | Buat-Menard and Arnold (1978) |
| | 1976 (June) | 32 | Buat-Menard and Arnold (1978) |
| | 1987 (July) | 1.2 | Andres et al. (1993) |
| | 1987 (July) | 0.24 | Andres et al. (1993) |
| | (Total) | 0.86 | Andres et al. (1993) |
| | (Bocca Nuova) | 18 | Aiuppa et al. (2003) |
| | 2001 (3 May) | 8.4 | Aiuppa et al. (2003) |
| | 2001 (18 May) | 9.3 | Aiuppa et al. (2003) |
| | 2001 (5 June) | 51 | Aiuppa et al. (2003) |
| | 2001 (29 June) | 5.8 | Aiuppa et al. (2003) |
| | 2001 (12 July) | 15 | Aiuppa et al. (2003) |
| | (Lava flow) | 1.1 | Aiuppa et al. (2003) |
| | 2001 (19 July) | 2.0 | Aiuppa et al. (2003) |
| Monti Carcassoni, Italy | 2001 (19 July) | 14 | Aiuppa et al. (2003) |
| | (Vent) | 0.25 | Calabrese et al. (2011) |
| | 2004–2007 | 0.24 | Kotra et al. (1983) |
| El Chichón, Mexico | - | 1.1 | Galindo et al. (1998) |
| Popocatépetl, Mexico | - | 0.12 | Suzuki (1964) |
| Nasudake, Japan | - | 0.24 | |

S3 Dummy aerosol emission and radius relationship

Table S3: AEROCOM I (Dentener et al., 2006) aerosol types and the corresponding effective radius.

| Aerosol Type | Effective radius (μm) |
|----------------------------------------------------|------------------------------------|
| Coarse sea salt | 2.50 |
| Coarse dust | 2.09 |
| Fine mode dust | 0.37 |
| Fine mode sea salt | 0.22 |
| Biomass burning-derived particulate organic matter | 0.095 |
| Biomass burning-derived black carbon | 0.095 |
| Biofuel-derived particulate organic matter | 0.095 |
| Biofuel-derived black carbon | 0.095 |
| Ultrafine mode sea salt | 0.037 |
| Fossil fuel-derived particulate organic matter | 0.036 |
| Fossil fuel-derived black carbon | 0.036 |

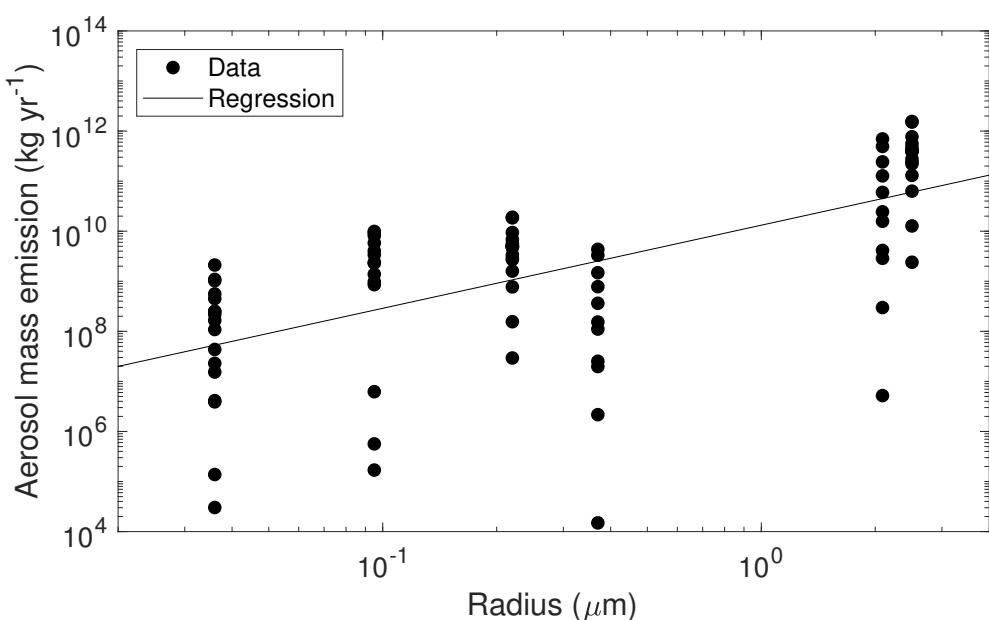


Figure S1: Relationship between aerosol radius and mass emissions in AEROCOM I inventory (Dentener et al., 2006). Emissions are aggregated to 10° latitude bands, to reflect the dummy latitude input parameter in this study.

S4 Validation of surrogate models

A set of 50 additional validation runs was created by enriching the existing training set so that a pseudo-Latin hypercube of 450 runs was formed (using the UQLAB function `uq_LHSify`). This ensures that our validation set is testing new regions of the parameter space compared to the training set.

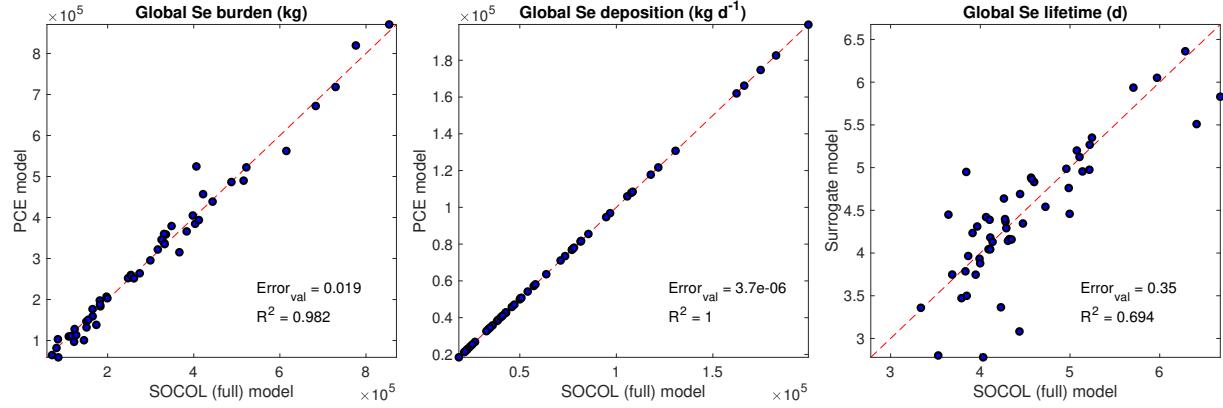


Figure S2: Comparison of SOCOL-AER results with surrogate model estimates for the 50 validation runs. Scatter plots and the one to one line are shown for the global Se burden (*left*), the global Se deposition flux (*center*), and the global Se lifetime (*right*), which is calculated by dividing the burden by deposition. The compared values are shown as blue circles and the 1:1 line as a red line.

In Fig. S2, we show the results of the validation runs compared to the predictions of the surrogate models for the global Se burden, deposition flux, and lifetime. The global lifetime surrogate model is calculated by dividing the PCE of the global burden by the PCE of the global deposition flux. The LOO error of the burden PCE is around 0.02 and the LOO of the deposition flux PCE is on the order of 10^{-6} . We can compare these LOO error to the validation errors, which are calculated from the independent validation set as (Marelli and Sudret, 2019):

$$\text{Error}_{\text{val}} = \frac{N-1}{N} \left[\frac{\sum_{i=1}^N \left(\mathcal{M}(\mathbf{x}_{\text{val}}^{(i)}) - \mathcal{M}^{\text{PCE}}(\mathbf{x}_{\text{val}}^{(i)}) \right)^2}{\sum_{i=1}^N \left(\mathcal{M}(\mathbf{x}_{\text{val}}^{(i)}) - \hat{\mu}_{Y_{\text{val}}} \right)^2} \right]$$

where N is the number of validation runs, $\mathcal{M}(\mathbf{x}_{\text{val}})$ is the SOCOL-AER model output of the validation runs, $\mathcal{M}^{\text{PCE}}(\mathbf{x}_{\text{val}})$ is the prediction of the surrogate model for the sample points in the validation set, and $\hat{\mu}_{Y_{\text{val}}}$ is the sample mean of validation set output.

The validation errors match the LOO errors for the global Se burden and Se deposition, showing that the LOO error is behaving correctly. However, the surrogate model for the global lifetime shows a relatively high validation error of around 0.35. The global selenium lifetime is clearly a difficult parameter to surrogate model with PCEs, likely requiring much more than 400 training

runs to be accurately modelled. The error of the surrogate model that we use is better than directly calculating a PCE for the atmospheric Se lifetime, which shows a validation error of 0.71.

In the second sensitivity analysis of the paper, we looked into the factors that affect Se deposition in each grid box. The validation dataset verifies the accuracy of the deposition flux surrogate models. We compare the simulated deposition in the 50 validation runs at all 8192 horizontal grid boxes with the PCE-predicted values, i.e. $50 \times 8192 = 409\,600$ points (Fig. S3). The left plot shows the results in linear space and the right plot in logarithmic space, since in the linear plot the smaller deposition fluxes collapse around 0. Most points fall around the 1:1 line, with the validation error being 0.013.

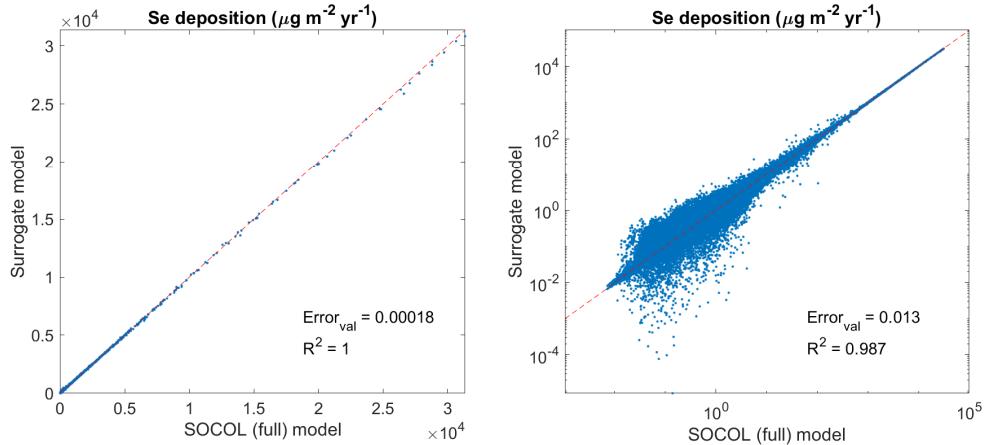


Figure S3: Comparison of SOCOL-AER results with surrogate model estimates for the 50 validation runs. Scatter plots and the one to one line are shown for Se deposition flux in every grid box on a linear scale (*left*) and a logarithmic scale (*right*). The compared values are shown as blue points and the 1:1 line as a red line.

S5 Leave-one-out error of PCE

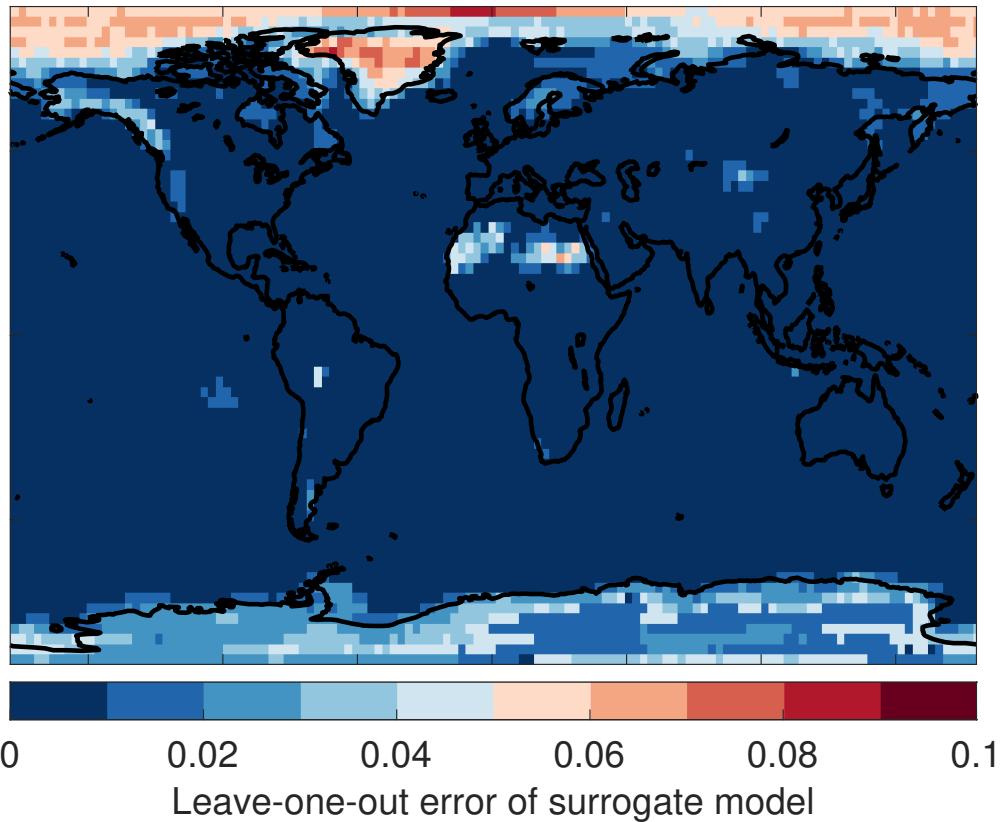


Figure S4: Leave-one-out error of surrogate models used to represent the total Se deposition fluxes from SOCOL-AER. The colorbar is chosen to highlight values that are above 0.05, which are shown in shades of red.

S6 References

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