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# Ambient Aerosol Exposure–Response as a Function of Particulate Surface Area: Reinterpretation of Historical Data Using Numerical Modelling

**A. D. MAYNARD[1]* and R. L. MAYNARD[2]**

[1]*National Institute of Occupational Safety and Health, Cincinnati, OH, USA;* [2]*Department of Health, London, UK*

**It has been hypothesized that the curvilinear response between British Smoke (BS) and excess mortality in London between 1958 and 1972 may be attributable to a linear response with respect to particulate number or surface area concentration. A numerical model has been developed and used to derive relationships between aerosol number, surface area and mass concentration under idealized environmental conditions. Modelling demonstrates that for a constant aerosol generation rate and rapid mixing, generalized functions can be derived that describe particle number versus mass concentration, and surface area versus mass concentration. The results indicate that the epidemiology data do not support a linear association between particle number concentration and mortality rate. However, a transformation between BS and particulate surface area is presented that leads to a linear association between aerosol surface area concentration and mortality rate. A critical mass concentration is defined, below which aerosol surface area varies linearly with mass. Above the critical mass concentration, numerical modelling supports the hypothesis that aerosol surface area is a more appropriate indicator of health effects associated with exposure.**

*Keywords:* exposure metrics; environmental exposure; aerosol surface area; numerical modelling

## INTRODUCTION

Although a number of epidemiology studies have indicated a linear relationship between ambient particulate mass and increased risk of mortality, historical data associating mortality with British Smoke (BS) indicate a negative deviation from a linear relationship at high mass concentrations (Schwartz and Marcus, 1990). It has been suggested that the non-linearity is attributable to the relationship between BS and particulate mass concentration. However, published data suggest the BS fraction of ambient aerosol is reduced at low mass concentrations, indicating that the mortality rate would show a greater negative deviation from a linear relationship at high mass concentrations if plotted against mass concentration. An alternative hypothesis, supported by a number of toxicology studies, is that mortality is related to aerosol number or surface area. The transformation between BS and these respective exposure metrics would require detailed information on the aerosol size distribution and particle shape. There are no contemporary experimental data that would allow such a transformation. However, it is possible that simplified modelling of the relationship between exposure metrics under appropriate conditions could provide sufficient information to indicate the validity of the proposed hypothesis. If it is assumed that BS is proportional to mass concentration, and that a linear exposure–response relationship exists between mortality and either particle number or surface area at low doses, then the exposure–response relationship seen in the Schwartz and Marcus data will reflect the form of the transformation between aerosol mass concentration and the most appropriate exposure metric. In this paper we use a simplified numerical model to estimate the temporal evolution of aerosol size distribution and concentration when generating into a defined volume at a constant aerosol generation rate. From the modelled distribution data, relationships between aerosol number, surface area and mass concentration are derived, and compared to the exposure–response relationship between mortality and BS.

*Author to whom correspondence should be addressed.
e-mail: amaynard@cdc.gov

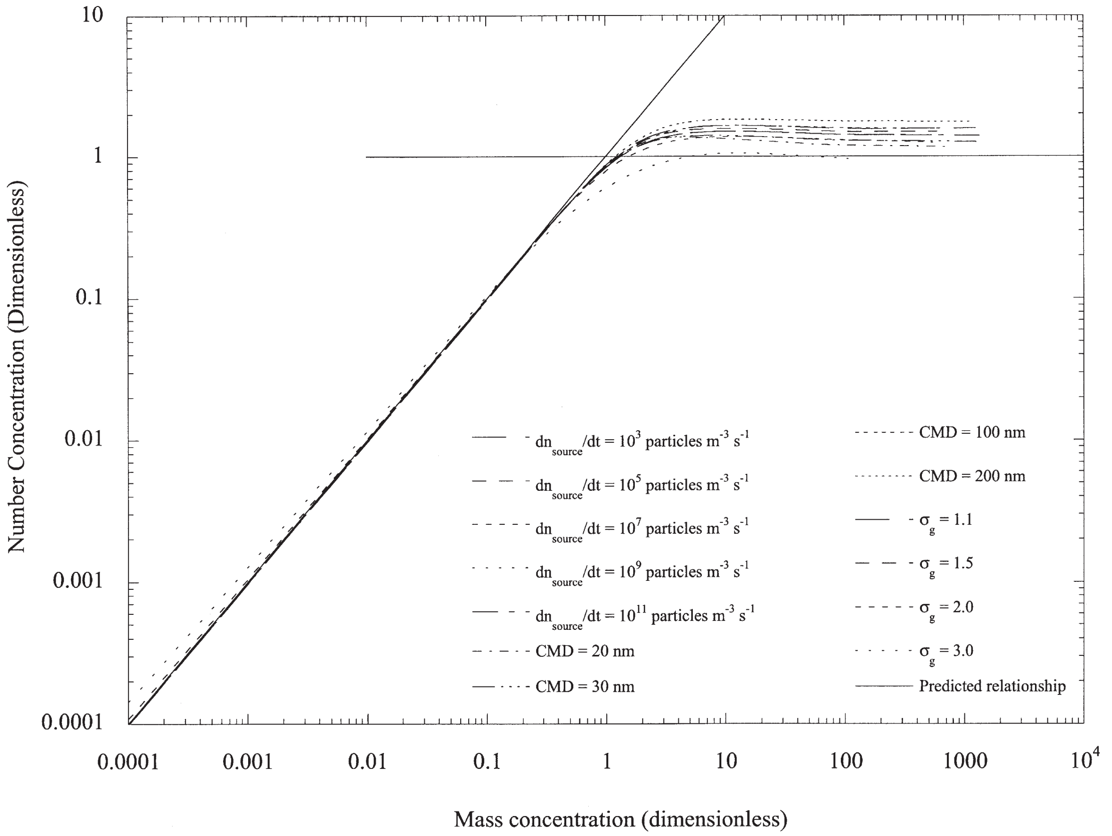


**Fig. 1.** Plotting modelled dimensionless number concentration against dimensionless mass concentration for a range of aerosol source parameters without gravitational settling. The reference source aerosol has $dn_{source}/dt = 10^7$ particles/m$^3$/s, $CMD = 50$ nm and $\sigma_g = 1.7$.

## MATERIALS AND METHODS

A simplified numerical model was developed to predict the temporal evolution of aerosol size distribution through generation, coagulation, gravitational settling and diffusional deposition. As the model was used to predict aerosol behaviour subsequent to initial particle formation and coagulation, condensational particle growth was not included. Coagulation was modelled using the Smoluchowski theory of coagulation, with the Fuchs coagulation coefficient correction factor was incorporated to allow interactions between particles from a few nanometers in diameter to tens of micrometers in diameter (Fuchs, 1964). The aerosol size distribution at time $t + \Delta t$ was estimated using successive estimates of the rate of change of number concentration as a function of particle diameter $d$ [$dn(d)/dt$] until changes in the estimated number concentration at time $t + \Delta t$ fell below a preset threshold (set to 1%). Mass was conserved during the modelling process, and the time steps and particle diameter bin widths used were dynamically modified to minimize computing time. The model was validated against experimental measurements of broad aerosol distributions with time in a sealed chamber with stirred settling, and shown to agree well with the experimental data.

To estimate the relationship between aerosol number ($n$), surface area ($s$) and mass ($m$) concentration in an idealized urban aerosol, a number of simplifying assumptions were made. At $t = 0$ the model was initialized with no background aerosol, and a single source generating a log-normal aerosol distribution characterized by its count median diameter ($CMD$), geometric standard deviation ($\sigma_g$) and generation rate ($dn_{source}/dt$). Spherical particles with a density $\rho$ of 2300 kg/m$^3$ (graphitic carbon) were assumed throughout the model. $CMD$ (of the generated aerosol), $\sigma_g$ and $dn_{source}/dt$ remained constant with time. The aerosol was generated into a cubic volume of 25 m$^3$, and rapid mixing was assumed. This volume was assumed to be surrounded by identical aerosol systems on all sides, and thus diffusional losses were ignored. The model was run both with no gravitational settling, and also with gravitational

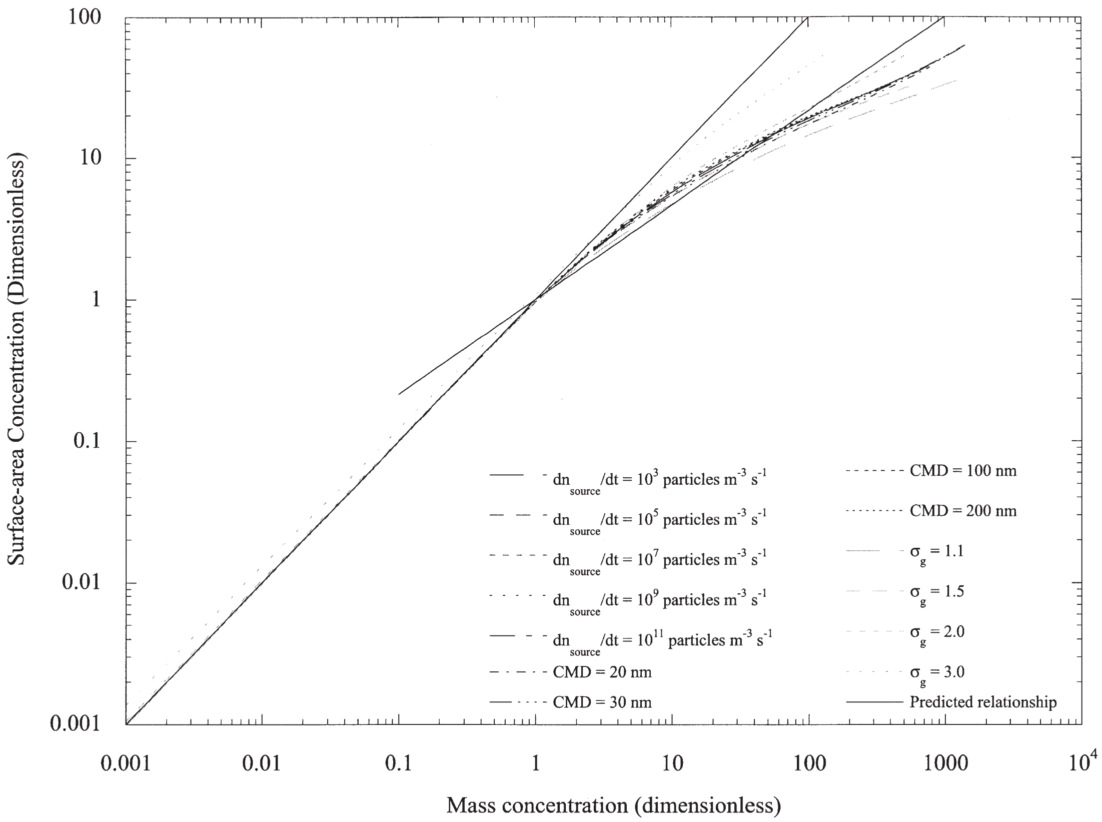


**Fig. 2.** Plotting modelled dimensionless surface area concentration against dimensionless mass concentration for a range of aerosol source parameters without gravitational settling. The reference source aerosol has $dn_{source}/dt = 10^7$ particles/m³/s, *CMD* = 50 nm and $\sigma_g = 1.7$.

settling included (in this case, assuming no flux of particles settling into the modelled volume from above). While representing an extremely simplified system, the model allowed the relationship between $n$, $s$ and $m$ to be followed with time.

Given the simplicity of the model, the relationships between $n$, $s$ and $m$ were dictated by the source aerosol (characterized by *CMD*, $\sigma_g$ and $dn_{source}/dt$) and time. The significance of each of the source terms was investigated by running the model with $dn_{source}/dt = 10^3$, $10^5$, $10^7$, $10^9$ and $10^{11}$ particles/m³/s (with $\sigma_g = 1.7$, *CMD* = 50 nm), *CMD* = 20, 30, 50, 100, 200 nm ($\sigma_g = 1.7$, $dn_{source}/dt = 10^7$ particles/m³/s) and $\sigma_g = 1.3$, 1.5, 1.7, 2.0, 3.0 ($dn_{source}/dt = 10^7$ particles/m³/s, *CMD* = 50 nm).

## THEORETICAL DERIVATION OF APPROXIMATING TRANSFORMATIONS

At low mass concentrations it can be assumed that the generation rate dominates coagulation and deposition rates, resulting in a linear relationship between $n$, $s$ and $m$. Assuming spherical particles,

$$n(m \ll m_{crit}) = \frac{6m}{\pi d_{\bar{m}}^3} \qquad (1)$$

$$s(m \ll m_{crit}) = \frac{d_{\bar{s}}^2}{d_{\bar{m}}^3}\frac{6m}{\rho}$$

where $d_{\bar{m}}$ and $d_{\bar{s}}$ are the diameters of average mass and surface area, respectively, within the source aerosol size distribution. $m_{crit}$ is a critical mass concentration defining low and high concentration regions. Above $m_{crit}$ the coagulation rate equals the generation rate (assuming no gravitational settling), leading to $n$ reaching a constant value of

$$n(m \gg m_{crit}) = \left(\frac{1}{K}\frac{dn_{source}}{dt}\right)^{\frac{1}{2}} \qquad (2)$$

where $K$ is the coagulation coefficient. $s$ will continue to rise if there are no depositional losses, giving

$$s(m \gg m_{crit}) = \pi d_{\bar{s}}^2\left(\frac{1}{K}\frac{dn_{source}}{dt}\right)^{\frac{1}{2}} \qquad (3)$$

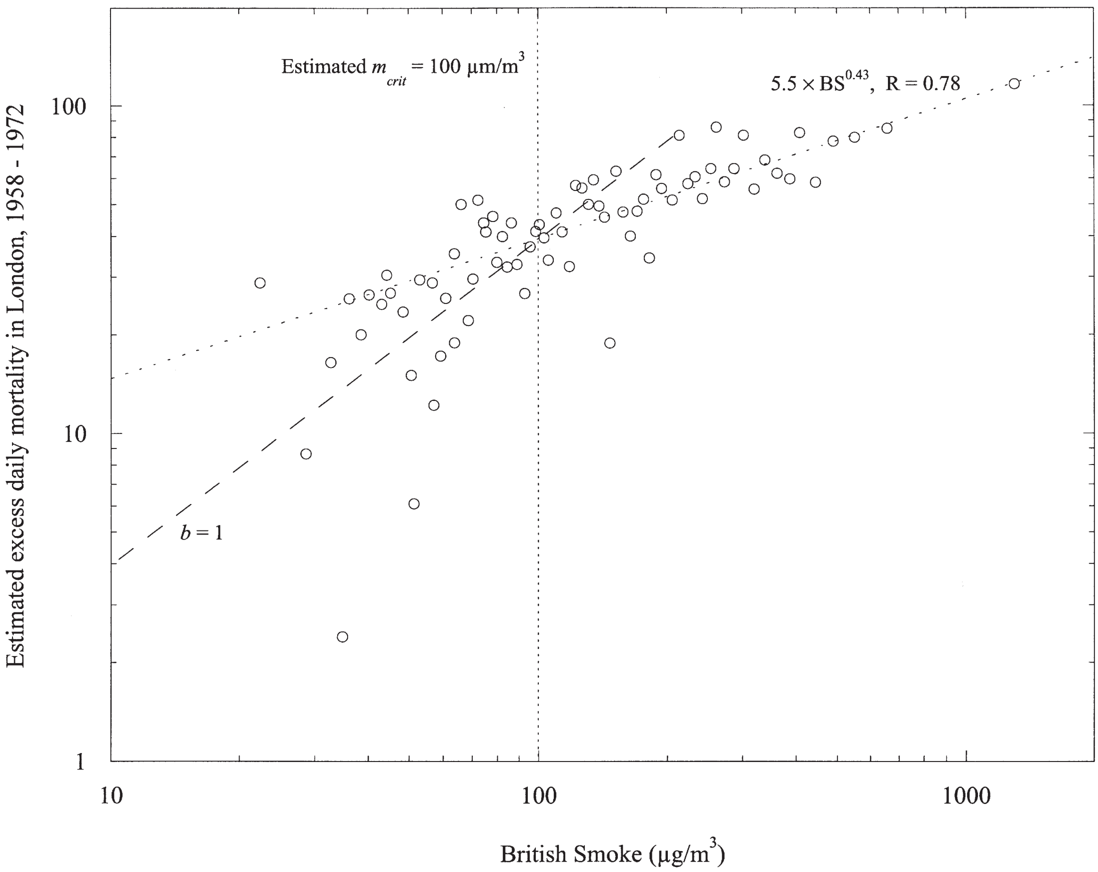


**Fig. 3.** Estimated excess daily mortality in London versus BS for the winters of 1958–72, derived from Schwartz and Marcus (1990), assuming a baseline mortality rate of 250 deaths/day. Data above 300 µg/m$^3$ have been fitted with a power function of the form $a\text{BS}^b$ using least-square regression.

In this case, $d_{\bar{s}}$ depends on the overall size distribution and is not determinable analytically. Assuming the overall aerosol size distribution is log-normal with a geometric standard deviation of $\sigma_g$ (aerosol) gives

$$s(m \gg m_{\text{crit}}) = \pi\left(\frac{6m}{\pi}\right)^{\frac{2}{3}}\left(\frac{1}{K}\frac{\mathrm{d}n_{\text{source}}}{\mathrm{d}t}\right)^{\frac{1}{6}} e^{-[\ln\sigma_g(\text{aerosol})]^2} \quad (4)$$

Thus, it is estimated that at high mass concentrations $s \propto m^{2/3}$. To simplify comparison of $n$, $s$ and $m$ under different conditions, each quantity can be made dimensionless using the following conversions:

$$n_{\text{dim}} = n\left(\frac{1}{K}\frac{\mathrm{d}n_{\text{source}}}{\mathrm{d}t}\right)^{-\frac{1}{2}} \quad (5)$$

$$s_{\text{dim}} = s\frac{1}{\pi d_{\bar{s}}^2}\left(\frac{1}{K}\frac{\mathrm{d}n_{\text{source}}}{\mathrm{d}t}\right)^{-\frac{1}{2}} \quad (6)$$

$$m_{\text{dim}} = m\frac{6}{\rho\pi d_{\bar{m}}^3}\left(\frac{1}{K}\frac{\mathrm{d}n_{\text{source}}}{\mathrm{d}t}\right)^{-\frac{1}{2}} \quad (7)$$

Using dimensionless concentrations, the critical dimensionless mass concentration separating low concentration and high concentration regions is $m_{\text{dim}}(\text{crit}) = 1$, and the maximum estimated dimensionless number concentration is $n_{\text{dim}} = 1$.

## RESULTS

Figures 1 and 2 show $n_{\text{dim}}$ and $s_{\text{dim}}$ versus $m_{\text{dim}}$, respectively, with no gravitational settling. Similar general trends were seen with gravitational settling included. Modelled relationships between $n$ and $m$ follow a similar form to the estimated relationship, with $n_{\text{dim}}$ tending to a value between 1 and 2 above $m_{\text{dim}}(\text{crit})$. The shape of neither the predicted nor the modelled curves indicates a suitable transformation between BS and particle number concentration that would result in a linear exposure–response relationship with respect to mortality. The modelled relation-

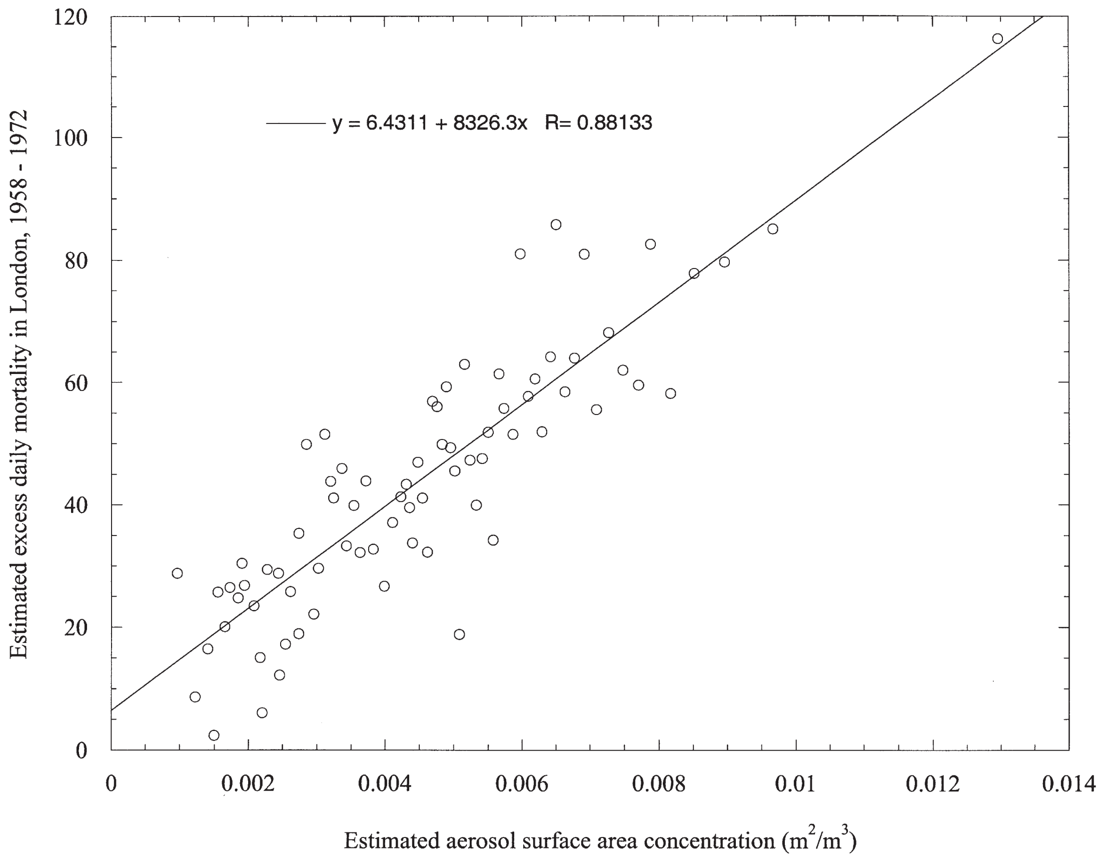


**Fig. 4.** Estimated excess daily mortality in London versus BS for the winters of 1958–72, derived from Schwartz and Marcus (1990), assuming a baseline mortality rate of 250 deaths/day. A source aerosol with *CMD* = 30 nm, $\sigma_g = 1.7$, $dn_{source}/dt = 8 \times 10^8$ particles/m$^3$/s has been assumed, derived from $m_{crit} = 100$ µg/m$^3$ and $b = 0.43$ (Fig. 3).

ship between $s_{dim}$ and $m_{dim}$ approximates to the predicted relationship, with some deviation between data sets above $m_{dim}$(crit). In general the relationship between surface area concentration and mass concentration above $m_{dim}$(crit) can be approximated by the function $s_{dim} = a \times m^b_{dim}$, with $b < 2/3$ ($a$ is a constant). There is evidence from the modelled data without gravitational settling that at high mass concentrations $b$ may approach the predicted value of 2/3, although the data do not extend to sufficiently high mass concentration values to confirm this.

## DISCUSSION

If it is assumed that excess mortality is proportional to $s_{dim}$, the numerical model indicates that it should also be proportional to $m^b_{dim}$, and thus $BS^b$. Values of $b$ for the modelled data vary with aerosol generation conditions and mass concentration, although they typically lie within the range $0.4 < b < 0.55$ (estimated). Figure 3 shows estimated excess mortality against BS, assuming a nominal baseline of 250 deaths/day. There is an indication of two regions within the data, with a critical BS concentration defining the regions at ~100 µg/m$^3$. Applying a least square regression to data at mass concentrations >300 µg/m$^3$ gives excess deaths proportional to $BS^{0.43}$ ($R = 0.78$). The value of $b$ of 0.43 is within the range of values estimated from the numerical model. Figure 4 shows daily deaths against the transform of BS to aerosol surface area concentration using appropriate values derived from the modelled data. Although this transformation is based on a number of highly simplifying assumptions, the resulting linear exposure–response relationship clearly supports the hypothesis that the curvilinear relationship between BS and mortality is indicative of excess deaths being more appropriately associated with aerosol surface area.

## CONCLUSIONS

In this study it has been hypothesized that the curvilinear exposure–response relationships between BS and mortality in London in the 1950s–1970s may be interpreted as a linear response between aerosol surface area or particle number concentration and excess mortality. Simplified numerical modelling has indicated that the relationship between aerosol mass and surface area concentration under conditions of constant aerosol generation is of a suitable form to transform the curvilinear response to a linear response. It has been estimated that under such conditions $s \propto m^b$ where $b$ typically lies between 0.4 and 0.55. Inspection of mortality data published by Schwartz and Marcus indicate a similar relationship between excess deaths and BS at high mass concentrations, with $b = 0.43$. Using the modelled data to estimate feasible aerosol generation rates and properties, a plausible transformation between BS and aerosol surface area has been indicated. As the transformation relies on many simplifying assumptions, it is only indicative of the actual concentration. However, the analysis strongly supports the hypothesis that response is related to particulate surface area, and has developed a framework within which estimates of surface area may be derived from historic mass concentration measurements.

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