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Space shuttle ice nuclei

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With the advent of the space shuttle era, increasing rocket activity is expected in the Earth's upper atmosphere. The space shuttle solid-fuelled booster rockets emit ~150 tonnes of alumina (Al_2O_3) particles per launch, much of which spreads throughout the upper troposphere and stratosphere¹. Such particles can seed aerosols and clouds, and might therefore cause changes in the terrestrial radiation balance and climate². Estimates are made here showing that average ice nuclei concentrations in the upper troposphere could increase by a factor of 2, and that an aluminium dust layer weighing up to 1,000 tonnes might eventually form in the lower stratosphere.

The morphology of alumina particles collected in high-altitude Titan exhaust trails may be summarized as follows^{1,3}: typically smooth and spherical with occasional deeply-grooved surface structures; hexagonal α - Al_2O_3 or cubic γ - Al_2O_3 crystallography; heavy-element composition dominated by Al with smaller amounts of K, Na, Ti, Fe and Si; trace quantities of chlorine adsorbed on surfaces; and densities in the range of ~1.5 to ~3.5 g cm^{-3} , suggesting individual particles with a porous or hollow construction.

Previous theoretical investigations⁴⁻⁶ focused on the role of Al_2O_3 particles as stratospheric condensation nuclei (CN, considered here as particles capable of nucleating supersaturated sulphuric acid vapour). However, space shuttle Al_2O_3 particles emitted in the stratosphere eventually filter downwards into the troposphere, after being widely dispersed over the hemisphere of injection. Inasmuch as alumina provides an excellent ice-forming epitaxy⁷, they might contribute significantly to the population of background ice deposition nuclei (IN) at high altitude. IN produce ice crystals directly from the vapour phase at low temperatures (-5° to -25°C) and supersaturations ($\geq 10\%$, or equivalently, relative humidities over ice ~100–110%). Cloud condensation nuclei (CCN), which can nucleate liquid water droplets at supersaturations $\leq 1\%$, are not subject to great influence by space shuttle emissions on a global scale.

Parungo and Allee^{8,9} and Hindman and co-workers¹⁰⁻¹³ studied IN concentrations in the stabilized ground clouds of solid-fuelled Titan rockets, which emit copious quantities of Al_2O_3 dust. They also discuss related laboratory data. Recognizing that the conditions in ground clouds are quite different from the conditions in high-altitude launch plumes, a consensus of the ground-cloud research nevertheless points to copious production of IN by rocket engines, perhaps as many as 10^{10} IN per gramme of fuel burned (active at -16° to -20°C and 101% relative humidity over ice).

To estimate the accumulation of space shuttle IN (or SSIN) in the upper troposphere and lower stratosphere, a comprehensive one-dimensional model of multicomponent atmospheric aerosols was used^{14,15}. The model segregates sulphate aerosols and un-nucleated CN particles, and keeps track of the amounts of different CN materials incorporated into the aerosols by nucleation and coagulation. In this manner, the Al_2O_3 component of aerosols in the size range from 0.001 to 10 μm radius (subdivided into 40 distinct size bins) was calculated for the size- and height-dependent Al_2O_3 particle deposition rate associated with the space shuttle. The aerosol physics and chemistry treated in the model are discussed elsewhere¹⁴. In the present simulations, micrometeorites, which are the dominant natural stratospheric particles above ~1 μm radius, are neglected; their presence does not materially alter the results.

The adopted Al_2O_3 mass deposition profile corresponds to 52 space shuttle launches per year, with emissions averaged over the Northern Hemisphere and smoothed in time to yield a continuous injection rate⁴. All of the aluminium burned was assumed to condense as dust. Particle interactions in the rocket wake were ignored, as these interactions act mainly to reduce the particle population below ~0.01 μm radius (see below). Al_2O_3 particle injection above 8 km was included in the calculations, the total mass deposition amounting to ~5,000 tonnes yr^{-1} . The size distribution of the injected particles (the number injected per cm^3 of air per μm radius) was taken to be constant below 0.01 μm radius, and to vary as $r^{-\alpha}$ above 0.01 μm . The latter dependence, with α in the range 3.5–4.5, is suggested by direct sampling in Titan exhaust plumes^{1,8,9}. The average particle density was assumed to be 1.7 g cm^{-3} , which emphasizes the number deposited.

The Al_2O_3 particle nucleation time in the stratosphere, τ_n , and the aerosol 'rainout and washout' lifetime in the troposphere, τ_r , were parameterized in a simple manner. For aluminium oxide dust, a fixed, relatively short nucleation time of 100 s was assumed as a base case¹⁶. Longer nucleation times up to 10^6 s were also tested. The simulated rainout loss rate decreases linearly from the ground-level value τ_r^{-1} to zero at the tropopause¹⁴. The baseline rainout rate of 10^{-6} s^{-1} at the ground translates into a rainout rate of $\sim 10^{-7} \text{ s}^{-1}$ near 10 km; ground-level rates as small as 10^{-7} s^{-1} were also considered.

To estimate the concentrations of SSIN in the upper troposphere and lower stratosphere using model predictions, three factors were taken into account: the composition of the particles, the extent of surface poisoning, and the size of the particles. The average composition of the sulphate aerosols in a given model size bin, i , is specified by the material volume fractions, $f_i(j)$, where j is a material index. Similarly, the volume fraction of material j in CN of size i is $f_i^{\text{CN}}(j)$. In the present work, the aerosol materials consist of three types: sulphuric acid aqueous solution, 'tropospheric' particulates, and aluminium oxide dust. The CN are composed of only the last

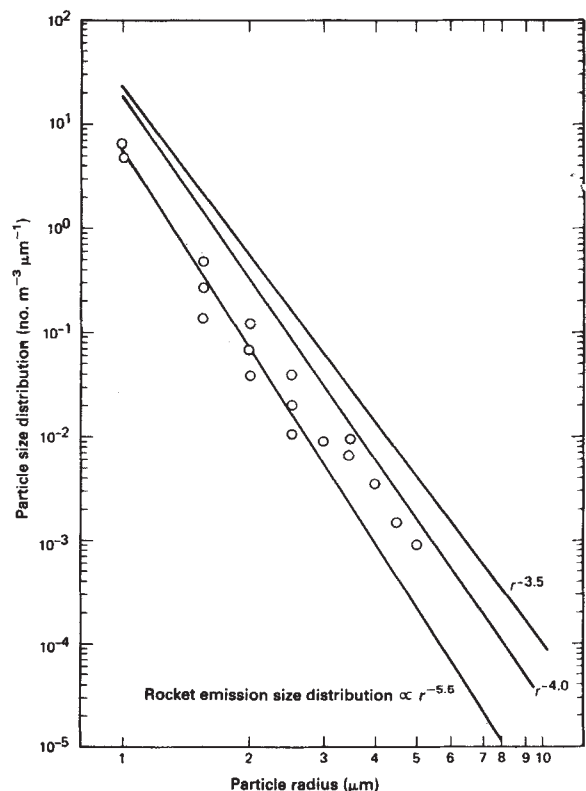


Fig. 1 Calculated stratospheric aerosol size distributions at 20 km with space shuttle particulate injection. 52 shuttle launches yr^{-1} , with emissions averaged over the Northern Hemisphere, were assumed. In each calculation, a nucleation time of 100 s was used (see text). Observations are compared: O, data from ref. 3 for Al_2O_3 at 20 km, multiplied by 10.

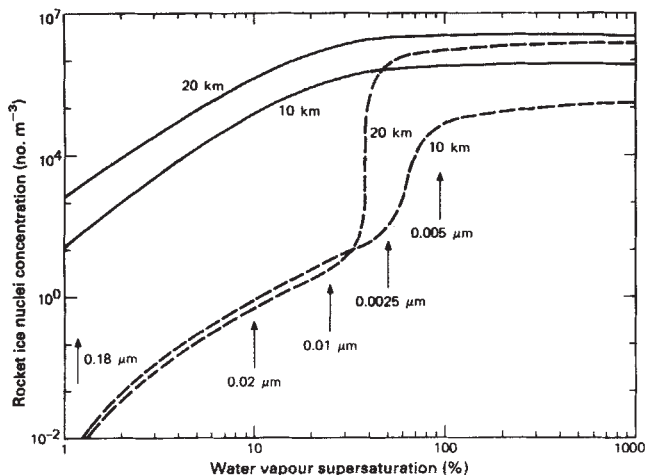


Fig. 2 Calculated space shuttle ice nuclei concentrations at 10 and 20 km altitude corresponding to different water vapour/ice supersaturations. The results were obtained for 52 shuttle launches per year with emissions averaged over the Northern Hemisphere, a dust emission size distribution of $r^{-4.5}$, and a nucleation time constant (τ_n) of 100 s. Estimates of the maximum (solid line) and average (dashed line) IN concentrations are given (see text). Also indicated are the minimum radii of spherical ice nuclei for several water vapour supersaturations at -30°C .

two materials. The maximum number of CN identified as active SSIN in each model size bin was taken as the fraction, $f_i^{\text{CN}}(\text{Al})$, of the total number of CN in that size bin. Nucleated CN are classified as acid aerosols in the model, and are discussed below. Possible deactivation of Al_2O_3 CN by coagulation with background 'tropospheric' CN was neglected in the stratosphere, as the background CN concentrations are so low ($<10\text{ cm}^{-3}$).

For H_2SO_4 aerosols, two estimates of the SSIN concentration were made (and added to the CN SSIN concentrations derived above). They are a 'maximum' number and an 'average' number. Such an approach was called for because the precise extent of the poisoning of individual alumina particles cannot be predicted with the model. Rather, the average sulphate and Al_2O_3 composition fractions of all the aerosols in a given model size bin are predicted.

The maximum concentration of (nearly) pure Al_2O_3 particles identifiable among the total population of aerosols in size bin i was first estimated as

$$n_i(\text{Al})_m = n_i f_i(\text{Al}) \quad (1)$$

where n_i is the concentration of aerosols of size i (number cm^{-3} , excluding CN), and $f_i(\text{Al})$ is the volume fraction of Al_2O_3 in aerosols of size i . However, the present model also provides information on the aerosol core size distribution in terms of the core volume moment fraction, h_i , which is a measure of the size dispersion of the cores in aerosols of size i , independent of core composition¹⁴. It can be shown that using h_i and f_i [where $f_i = \sum_j f_i(j)$], the maximum number of essentially-pure core aerosols may also be approximated as

$$n_i(\text{core})_m \doteq n_i [h_i^2 - f_i^2] / [1 + h_i^2 - 2f_i] \quad (2)$$

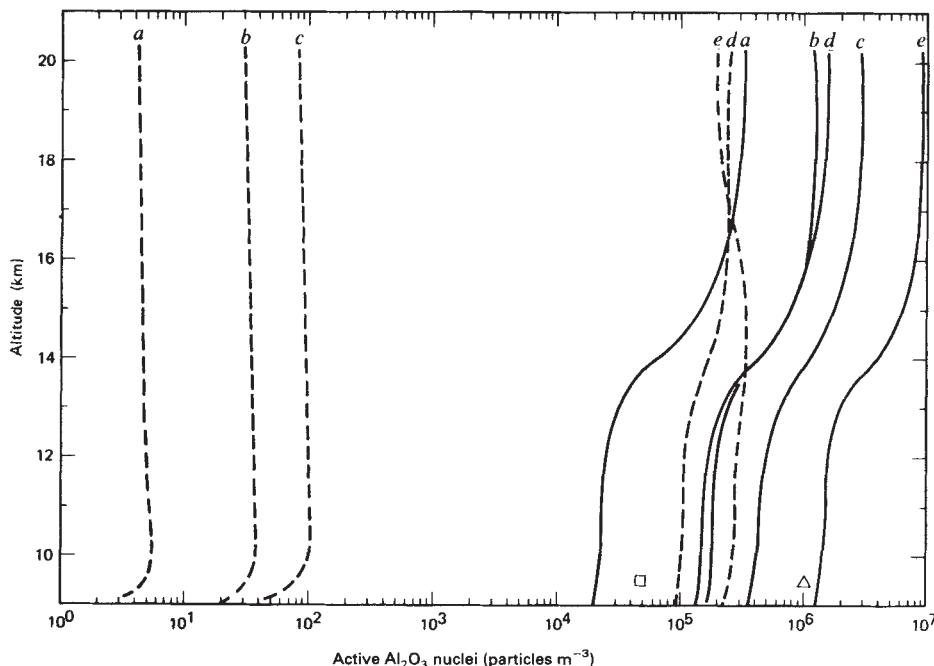
Accordingly, the maximum concentration of aerosol SSIN of size i was taken as the lesser of $n_i(\text{Al})_m$ and $n_i(\text{core})_m$.

The 'average' number of aerosol SSIN was calculated by assuming that all of the aerosols of size i have an average coating, or film, of contaminating material (mainly liquid sulphuric acid). If, in any size bin, the contamination amounted to more than a uniform monolayer coating $\approx 4\text{ \AA}$ thick, all of the aerosols in that bin were considered to be poisoned.

The effect of particle size on the activity of rocket nuclei was estimated in two ways. In one case, the SSIN concentrations deduced above (including the contributions of CN and droplets) were truncated to exclude particles $<0.01\text{ }\mu\text{m}$ radius; such particles generally require supersaturations exceeding $\sim 10\%$ to cause nucleation at $\sim -20^\circ$ to -30°C (refs 16, 17). In the second approach, the effect of particle size on the water vapour pressures of small ice spheres was calculated using the Kelvin relation¹⁷. At a temperature of -30°C (adopted to estimate the critical size at each altitude) the minimum nuclei dimensions corresponding to water vapor supersaturations of 1, 10, 50, 100 and 900% are 0.19, 0.020, 0.0046, 0.0027 and 0.0008- μm radius, respectively.

Figure 1 shows projected aerosol size distributions at 20 km for several different Al_2O_3 emission size spectra, $r^{-\alpha}$. In the size range 1–10 μm , the aerosols are essentially pure alumina ($>95\%$ pure). The data of Brownlee *et al.*³ are plotted for comparison. The close agreement between the observed size distribution and the computational results suggests that the present shuttle simulations are physically reasonable. Brownlee *et al.*³ estimated the global stratospheric burden of Al_2O_3 in 1974–5 as ~ 100 tonnes. The stratospheric lifetime of particles in the size range 1–5 μm is ~ 1 month, implying an Al_2O_3 mass input rate of $\sim 1,000$ tonnes yr^{-1} worldwide. The space shuttle calculations in Fig. 1 yield ~ 10 times as many rocket alumina particles for a hemispherical input rate of $\sim 5,000$ tonnes yr^{-1} , in clear accord with the Brownlee *et al.* observations. The

Fig. 3 Predicted space shuttle ice nuclei (SSIN) concentrations in the lower stratosphere and upper troposphere. The cases shown are: a, $r^{-3.5}$, $\tau_n = 100\text{ s}$, $\tau_r = 10^6\text{ s}$; b, r^{-4} , $\tau_n = 100\text{ s}$, $\tau_r = 10^6\text{ s}$; c, $r^{-4.5}$, $\tau_n = 100\text{ s}$, $\tau_r = 10^6\text{ s}$; d, r^{-4} , $\tau_n = 10^6\text{ s}$, $\tau_r = 10^7\text{ s}$; e, r^{-4} , $\tau_n = 10^5\text{ s}$, $\tau_r = 10^6\text{ s}$ (see the text for an explanation of these parameters). In cases a–d, 52 shuttle launches per year were assumed, with the effluent spread uniformly over the Northern Hemisphere. In case e, a 10-fold increase in the numbers of launches was assumed. A minimum nuclei radius of $0.01\text{ }\mu\text{m}$ was used to estimate the SSIN abundances. Maximum (solid line) and average (dashed line) numbers of Al_2O_3 nuclei are shown. Δ , Observed ice nuclei concentration in cirrus clouds at -30°C . \square , Maximum ice crystal concentration in cirrus clouds.



corresponding predicted global mass of the stratospheric alumina layer is $\sim 1,000$ tonnes.

The computed size distributions in Fig. 1 have a relatively simple explanation. If rocket particles are injected at all altitudes with the same size distribution ($r^{-\alpha}$), and if particle growth by condensation and coagulation can be neglected, then the resultant atmospheric aerosol size distribution depends on radius roughly as $r^{-\alpha-\beta}$, where β is the exponent of the radius dependence of the particle fall velocity; that is, $v(r) \propto r^\beta$. In the lower stratosphere, $1 < \beta < 2$ for particles with radii in the size range 1–10 μm . Thus, for Al_2O_3 rocket particles one expects $5 \leq (\alpha + \beta) \leq 6$, as is found in Fig. 1.

Figure 2 illustrates the influence of water vapour supersaturation on apparent SSIN concentrations. As expected, higher supersaturations yield larger IN abundances. The abrupt increase in the 'average' IN curves at S values of 40–80% is due to a substantial number of nearly-pure rocket dust particles at sizes below $\sim 0.01 \mu\text{m}$. For meteorologically significant supersaturations ($< 10\%$), the average number of IN in Fig. 2 is nearly four orders of magnitude smaller than the maximum number of IN. The difference represents, in one sense, the uncertainty in predictions of rocket IN abundances.

Predicted SSIN concentrations in the upper troposphere and lower stratosphere, for various physical parameter values, are given in Fig. 3. Several trends in the data are noteworthy. As in Fig. 2, with fast Al_2O_3 nucleation ($\tau_n = 100$ s), there is a difference of a factor of 10^4 between the 'average' and 'maximum' numbers of SSIN. Obviously, there is ample sulphate material present in the atmosphere to contaminate most of the injected dust grains. The maximum SSIN concentrations would apply if Al_2O_3 particle surfaces were not efficiently nucleated and poisoned by H_2SO_4 vapour (see, for example, cases *d* and *e* in Fig. 3). Comparison of cases *b*, *d* and *e* suggests that the average SSIN concentration is roughly proportional to τ_n^{-1} . In other words, once nucleated, most Al_2O_3 grains accumulate enough sulphur vapour to be poisoned (in the model). Rainout has a much smaller influence on SSIN concentrations; the residence time of injected particles in the upper troposphere is controlled primarily by vertical diffusion (for the cases studied).

One might expect a short nucleation time ($\tau_n \sim 100$ s) to apply to Al_2O_3 dust released in the upper atmosphere. For one thing, the H_2SO_4 supersaturation ratio between 10 and 20 km altitude is quite large, $\sim 10^4$ – 10^5 (ref. 16). Hence, adsorption of H_2SO_4 on alumina surfaces should commence immediately on injection. Continuing growth of the dust particles larger than the critical (Kelvin) size would also be guaranteed. However, a factor which causes the effective nucleation time to be much longer is the time required to deposit a monolayer of acid molecules on the dust grain surfaces. The ambient quantity of H_2SO_4 vapour available in the neighbourhood of the launch plume is insufficient to coat more than a few per cent of the rocket particles. Nevertheless, it may be demonstrated that, within a day or so, individual launch plumes are dispersed widely enough for extensive acid adsorption to occur. This does not take into consideration the HCl vapour in the early launch plume, which acts to poison the dust surfaces at high concentrations^{9–11}, but appears to activate the surfaces at low concentrations¹³. Accordingly, an effective dust nucleation time $\leq 10^5$ s appears to be most appropriate for stratospheric conditions.

Many of the SSIN residing in the upper troposphere are injected between 8 and 12 km. At these altitudes, the transient space shuttle launch clouds should be considered. Case *e* in Fig. 3, which corresponds to a 10-fold enhancement in the average shuttle particle deposition rate, might crudely forecast the elevated IN concentrations to be expected in isolated launch plumes.

The maximum number of rocket IN that could be generated from 1 g of fuel, based on an r^{-4} Al_2O_3 dust size distribution and a minimum activation radius of 0.01 μm , is $\sim 10^{15}$ IN g^{-1} . This number corresponds closely to the maximum number predicted by the model. By contrast, Hindman *et al.*¹² measured

$\sim 10^{10}$ IN g^{-1} in small shuttle propellant burns, while the present calculations yield estimates of average long-term shuttle IN yields $\sim 10^{11}$ IN g^{-1} . Hence, the maximum SSIN profiles in Fig. 3 are very likely to represent upper-limit global-scale concentrations of artificial ice nuclei produced by space shuttles.

Typical IN concentrations measured at cirrus cloud heights are indicated in Fig. 3 (ref. 17). Maximum concentrations of ice crystals observed in cirrus clouds are also shown. Based on a comparison of measured and predicted IN concentrations, and the previous discussion, we conclude that planned space shuttle activity probably will not increase the global abundance of ice nuclei at cirrus cloud levels by more than a factor of 2, although larger transient increases might occur locally. Injected space shuttle alumina particles, on the other hand, could enhance the concentrations of stratospheric aerosols in the size range 1–10 μm radius by a factor of 10 or more, creating in the stratosphere an aluminium oxide layer exceeding 1,000 tonnes in weight.

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The Earth's early hydrosphere

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The date at which the Earth's hydrosphere came into existence is unknown. Although all current geological models for the very early Precambrian predict or suppose a hydrosphere^{1–4}, there are difficulties in reconciling this with geological and astronomical theories of planetary formation. Furthermore the effect of a substantial hydrosphere on the Precambrian climatic environment is difficult to determine. Recent developments make it possible to synthesize a picture of this environment during the 700 Myr preceding the deposition of the oldest known sediments⁵. We attempt such a synthesis here, drawing together work on the dynamics of accretion from the proto-planetary nebula^{6,7} and of planetary differentiation⁷, on the role of volatiles in impact cratering^{8–10}, and on the composition^{11–22}, isotope systematics^{23–25} and radiation climatology^{26–29} of the early hydrosphere. We can reconcile these diverse contributions if the hydrosphere—the inventory of excess volatiles¹—is allowed to differentiate during accretion, with most of the H_2O and CO_2 going rapidly into oceans and sediments. Both gradual outgassing³⁰ and a massive CO_2 atmosphere² are unlikely, and a reduced mantle beneath a neutral hydrosphere is not paradoxical^{1,3,17}. Results from both a one-dimensional radiative-convective (1D RC) model and a general circulation model suggest that self-regulating mechanisms are important in the climate of this early hydrosphere.