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June 1976

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ON SEEDING THE D REGION WITH CESIUM

By: J. W. CHAMBERLAIN

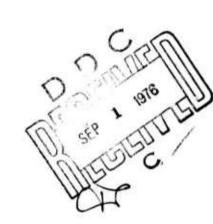
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20 ABSTRACT (Continued)

daytime release. Although the ionization will decrease rapidly at night, it will reappear again at sunrise, with electron densities of the order of 10³ cm⁻³ persisting for days. Disturbed radio propagation in VHF, and especially HF, would be present for days, possibly over distances up to several thousand kilometers. Seeding the D region with substantial quantities of cesium would provide a valuable diagnostic tool for investigation of the local atmospheric dynamics, but it could also produce undesirable effects on normal radio communications.

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I INTRODUCTION

Seeding the upper atmosphere with an alkali metal (sodium) was first proposed as an experiment by D. R. Bates. The objective was then to clarify certain aspects of the airglow chemistry, and although the subsequent experiments did little to further one's understanding of the airglow, seeding experiments became a major program in their own right. While some seeding materials were chosen with little apparent rationale while some seeding materials were chosen with little apparent rationale beyond an expensive curiosity, others (notably barium) have been widely used for the insight they provided to dynamics of the neutral and ionized upper atmosphere. Barium is readily ionized by unattenuated sunlight (IP = 5.2 eV) and Ba⁺ has the same electron configuration (5 p⁻⁶ 6s $^{-8}$ S_{1/2}) as cesium.

Cesium in moderate amounts (10 kg) has been seeded at 70, 80, and 90 km by the Air Force Cambridge Research Center and has been found to produce ion clouds in the daytime, except possibly at the lower altitude where chemical consumption may have prevented the creation of Cs .

We propose here that cesium should be further considered as a possible seed for the upper mesosphere or D region. Cesium is superior to any other seeding substance in that it will be largely ionized at the lowest altitudes of the natural ionosphere and probably has a long local lifetime against chemical destruction.

II INJECTION AND SPREADING

Suppose for definiteness that 7500 moles (one ton) of cesium (mass 133) is initially injected explosively from a point source at 80 km, where the ambient density is N \approx 5 \times 10 14 cm $^{-3}$. The cesium source is $\eta = 4.5 \times 10^{27}$ atoms. The initial expansion will reach a minimum radius given by

$$\frac{4}{3} \pi r^3 \sim \frac{\eta}{N} = \frac{4.5 \times 10^{27}}{5 \times 10^{14}}$$

or $r \sim 10^4$ cm before the Cs density equilibrates with the ambient atmosphere. We shall suppose in the following that the ejection extends vertically over a range of 10 km. The horizontal winds and turbulence will spread material at characteristic velocities of 10 to 100 m/s; hence within mirates the cloud must subtend a radius of several kilometers. After one day, with a horizontal eddy-diffusion coefficient of $K \sim 3 \times 10^{10}$ cm²/s, the cesium cloud would cover a radius of $r \sim \sqrt{Kt} = 500$ km.

There is considerable uncertainty in the horizontal eddy-diffusion coefficient. Certainly horizontal mixing is much faster than vertical mixing. The tentative value of K adopted here is suggested by the characteristic velocity differences (up to 150 m/s) seen over distances of 10 km in some of the early seeding experiments in the 80-to-100-km region. In addition, zonal winds could possibly transport the cloud several hundred or even thousand kilometers per day, but little is known of systematic winds in the D region.

Vertical motions are much slower. Eventually vertical mixing will destroy the effectiveness of the cesium by removing it from the D region, but that process would require many days or even weeks. The vertical motions are dominated in this region by turbulent mixing with an eddy coefficient of the order of $K_z \approx 5 \times 10^5$ cm /s. While its rate is rather uncertain, this large-scale mixing is much more important than molecular diffusion. Cesium will have a molecular diffusion coefficient of $D \sim 10^4$ cm /s that will produce vertical diffusion velocities, $D/\Delta z$, of only 10^{-2} cm/s. By contrast, the eddy mixing will proceed a factor of 50 faster, or at about 1/2 km/day.

In summary, the later-time motions consist of a slow vertical mixing, a moderate horizontal attenuation by turbulence, and an unknown but possibly rapid zonal transport. The thickness Δz , horizontal radius r, and mean density $\langle N \rangle$, are summarized at three time intervals in Table 1.

Table 1
ASSUMED SPREADING OF A ONE-TON CESIUM CLOUD

t	Δz (km)	r (km)	(N) (cm ⁻³)							
1/4 hr	10	50	6 × 10 ⁷							
1 day	10	500	6 × 10 ⁵							
4 days	10	1000	1.5×10^5							

III CHEMISTRY

Atomic Cs may be oxidized by one or more of the following:

$$Cs + O_3 \rightarrow CsO + O_2$$
, + 2.1 eV (1)

$$Cs + O + M \rightarrow CsO + M, + 3.2 \text{ eV}$$
 (2)

and

$$Cs + O_2 + M \rightarrow CsO_2 + M \qquad . \tag{3}$$

The principal reduction mechanism is likely to be

$$CsO + O \rightarrow Cs + O_2$$
, + 1.9 eV . (4)

For Cso_2 formed by reaction (3), the reduction cycle is probably

$$CsO_2 + O \rightarrow CsO + O_2 \tag{5}$$

followed by reaction (4).

Hydroxyl chemistry could complicate the cycles with CsH formed in place of CsO and

$$CsH + O \rightarrow Cs + OH$$
 (6)

replacing reaction (4). Similarly, CsO_2 could be removed by

$$CsO_2 + H$$
 or $CsO + OH$. (7a)

with the large amounts of ionization present (see Section IV), the chemistry may become further complicated by reactions between positive and negative ions. A quantitative discussion is hampered by a lack of information on reaction rates and even dissociation energies. The tentative value of 3.2 eV for dissociation of CsO is based on a theoretical calculation by Gusarov and Gorokhov. The estimated error is ±0.25 eV, but measurements on LiO and NaO indicate that the theoretical computations tend to be slightly larger than the experimental values.

With the postulated large amounts of cesium deposited, reaction (1) will be initially important in destroying 0_3 locally, and 0_3 reactions can consequently be omitted from the quasi-equilibrium chemistry, at least for the present qualitative discussion.

The principal oxidation process is probably reaction (3). The rate coefficient has been measured 7 at 2.1×10^{-33} cm s $^{-1}$ in the temperature range 1420 to 1600 °K, where it has twice the rate for the equivalent reaction with Na. However, the Na rate increases with decreasing temperature and is probably about 10^{-29} cm s $^{-1}$ at 150 °K; a similar value should be appropriate for reaction (3).

The portion of free Cs will then be

$$\frac{\text{Cs}}{[\text{CsO} + \text{CsO}_2]} = \frac{[0] k_4 k_5}{[0_2][\text{M}] k_3 (k_4 \cdot k_5)}$$

For illustration, if at 80 km, $= 5 \times 10^{14} \text{ cm}^{-3}$, $[0_2] = 10^{14} \text{ cm}^{-3}$, $[0] = 10^{11} \text{ cm}^{-3}$, and $k_4 \sim k_5 \sim 3 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$, we have

$$\frac{[\operatorname{Cs}]}{[\operatorname{CsO} + \operatorname{CsO}_2]} \sim 3 \qquad ,$$

or most of the Cs remains free at 80 km.

At appreciably lower altitudes, Cs would be chemically consumed and the high ionization levels possible at 80 km would not occur. Although based on plausibility values for \mathbf{k}_4 and \mathbf{k}_5 , this qualitative result is compatible with the earlier cesium seeding experiments producing ionization at 80 km but not in detectable amounts at 70 km.

Further, since the cesium chemistry probably parallels closely that for sodium, this result is compatible with Na twilight airglow observations, which indicate that free Na peaks at about 85 km and diminishes rather abruptly below 80 km. In this regard it is interesting to note that reaction (4) is energetically capable of producing $Cs(^2P)$ levels, which are the upper levels for the resonance lines, 8943 Å and 8521 Å. The Na "D" lines (5896 Å and 5890 Å) are probably produced by the analogous sodium reaction in the night airglow.

IV IONIZATION

The low ionization potential of 3.894 eV (3183 Å) for Cs will cause it to be ionized by solar radiation at comparatively low altitudes. With an overhead O_3 abundance above 80 km of 10^{14} molecules/cm², and an absorption cross section of σ (O_3 | 3200 Å) \approx 3 \times 10⁻²⁰ cm², the Cs deposited in this region will not be seriously shielded from sunlight. The Cs photoionization cross section near the threshold is $\sigma \approx 10^{-19}$ cm². With an average solar flux of 10^{13} photon/cm²-s-Å and a region of the order of 1000 Å wide free of attenuation of O_3 , the ionization rate is

$$\frac{d[Cs]}{[Cs] dt} = -10^{-3} s^{-1}$$

or an ionization time of about 1/4 hour.

Because of the low ionization potential (3.9 eV) of Cs, chargetransfer reactions of Cs⁺ with neutrals can be ignored. The ionization balance of cesium will be governed by mutual neutralization,

$$Cs^{+} + X^{-} \rightarrow Cs + X \qquad . \tag{8}$$

The principal factors in the ionization balance are the following. Free electrons are produced by photoionization of Cs and by photodetachment and associative detachment from D-region negative ions, such as 0^- , 0^-_2 , and the so-called "terminal" negative ion, still not firmly identified. Electrons are lost by radiative and three-body attachment. In addition, equilibrium for each negative ion is established by attachment, detachment, and mutual neutralization.

An approximate solution of the daytime equilibrium equations gives an ionization density of

$$[Cs^{+}] \approx [e] = 5 \times 10^{7} \text{ cm}^{-3}$$
,

a negative ion ratio of

$$\lambda \equiv \frac{[X]}{[e]} = 1.6 \times 10^{-4} \quad ,$$

and a cesium ion/neutral ratio of

$$\frac{[Cs^+]}{[Cs]} \sim 1$$
 .

An electron density of 3×10^7 cm $^{-3}$, which can be expected in the early period (see Table 1), corresponds to a critical frequency of about 50 MHz/s. The attenuation for vertical incidence over the 10-km perturbed region is about 4 dB at 400 MHz and 25 dB at 150 MHz. At later times, densities of N $^{\sim}$ 10 5 cm $^{-3}$ in the D region could cause long-lived daytime HF absorption over areas hundreds or even thousands of kilometers in radius.

V DISCUSSION

Cesium is especially suited as a seeding material for producing long-lived effects, and it could be traced for days or weeks in suitable abundances. It can exist in the free-ion form at low D-region altitudes probably better than any other atom. It has a low ionization potential, being photoionized by the solar flux below 3200 Å. Although K and Rb also have fairly low ionization potentials, their photoionization cross sections are not so favorable.

Suppose a cloud of cesium were deposited in the 80-km region. While zonal winds may carry it in a way still not known, the cloud will disperse only gradually. Each evening the ionization will largely disappear, only to reappear at sunrise.

The ions in the D region may also be subject to drifts induced by ionospheric electric fields, as is the case for barium ions released at higher altitudes. Very little is known about horizontal motions in this region, and a long-lived experiment—one in which the seed material can be tracked for days—would be desirable.

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