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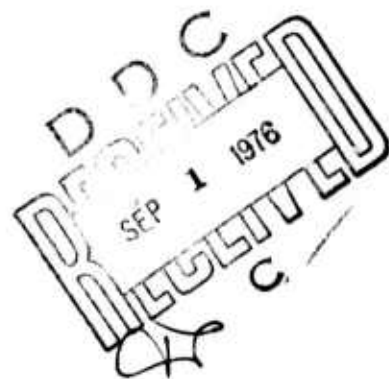
Technical Report JSR-74-5

June 1976

ON SEEDING THE D REGION WITH CESIUM

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Contract No. DAHC15-73-C-0370
ARPA Order No. 2504
Program Code No. 3K10
Date of Contract: 2 April 1973
Contract Expiration Date: 30 August 1976
Amount of Contract: \$2,062,014



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REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER JSR 74-5	2. GOVT ACCESSION NO.	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) ON SEEDING THE D-REGION WITH CESIUM	5. TYPE OF REPORT & PERIOD COVERED Technical Note	
6. AUTHOR J. W. Chamberlain	6. PERFORMING ORG. REPORT NUMBER SRI Project 3000	
7. PERFORMING ORGANIZATION NAME AND ADDRESS Stanford Research Institute Menlo Park, California 94025	8. CONTRACT OR GRANT NUMBER DAHC15-73-C-0370	
9. CONTROLLING OFFICE NAME AND ADDRESS Defense Advanced Research Projects Agency 1400 Wilson Boulevard Arlington, Virginia 22209	10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS ✓ ARPA Order-2504	
11. MONITORING AGENCY NAME & ADDRESS (if diff. from Controlling Office) SRI-JSR-74-5	12. REPORT DATE June 1976	13. NO. OF PAGES 17
14. DISTRIBUTION STATEMENT (of this report) Approved for public release; distribution unlimited. SRI-3000		15. SECURITY CLASS. (of this report) Unclassified (12) 15p.
15a. DECLASSIFICATION/DOWNGRADING SCHEDULE		
16. DISTRIBUTION STATEMENT for the abstract entered in Block 20, if different from report		
17. SUPPLEMENTARY NOTES		
18. KEY WORDS (Continue on reverse side if necessary and identify by block number)		
D Region	Daytime	Theoretical/experimental
Chemical reactions	Night	Diagnostics
Dynamics of	VHF, HF	Disturbances 6×10 to the 7th power/cc
Cesium seeding 1000	Radio probing	Radio communications
19. ABSTRACT (Continue on reverse side if necessary and identify by block number)		
If 10^3 kg (7500 moles) of cesium were deposited over a 10-km height interval centered about 80-km altitude, the cloud would spread within 1/4 hour to a 50-km radius, leaving a mean density of $[Cs] = 6 \times 10^7 \text{ cm}^{-3}$. Chemical reactions with oxygen would consume about 25% of the cesium, but most of it would remain in atomic form. Solar ionization is very effective, and Cs^+ will not transfer its charge to atmospheric constituents, which leads to nearly 100% ionization of the free Cs and to free-electron densities of nearly $3 \times 10^7 \text{ cm}^{-3}$ in less than an hour after a		

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19. KEY WORDS (Continued)

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daytime release. Although the ionization will decrease rapidly at night, it will reappear again at sunrise, with electron densities of the order of 10^5 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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CONTENTS

I	INTRODUCTION	1
II	INJECTION AND SPREADING.	2
III	CHEMISTRY.	4
IV	IONIZATION	7
V	DISCUSSION	9
	REFERENCES	10
	DISTRIBUTION LIST.	11

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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.¹ 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 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 ($5p^6 6s^2 S_{1/2}$) as cesium.

Cesium in moderate amounts (10 kg) has been seeded at 70, 80, and 90 km by the Air Force Cambridge Research Center² 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 $N \approx 5 \times 10^{14} \text{ cm}^{-3}$. 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 $r \sim 10^4 \text{ cm}$ 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 minutes the cloud must subtend a radius of several kilometers. After one day, with a horizontal eddy-diffusion coefficient of $K \sim 3 \times 10^{10} \text{ cm}^2/\text{s}$, the cesium cloud would cover a radius of $r \sim \sqrt{Kt} = 500 \text{ km}$.

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.³ 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 coefficient⁴ of the order of $K_z \approx 5 \times 10^5 \text{ cm}^2/\text{s}$. While its rate is rather uncertain, this large-scale mixing is much more important than molecular diffusion. Cesium will have a molecular diffusion coefficient⁵ of $D \sim 10^4 \text{ cm}^2/\text{s}$ 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 $1/2 \text{ km/day}$.

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.

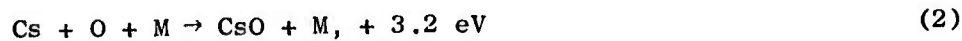
Table 1

ASSUMED SPREADING OF A ONE-TON CESIUM CLOUD

t	Δz (km)	r (km)	$\langle N \rangle$ (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:



and



The principal reduction mechanism is likely to be



For CsO_2 formed by reaction (3), the reduction cycle is probably

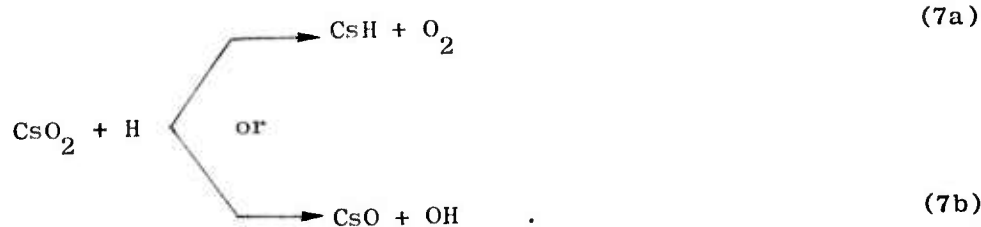


followed by reaction (4).

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



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



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.⁶ 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 O_3 locally, and O_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 measured⁷ at $2.1 \times 10^{-33} \text{ cm}^6 \text{ s}^{-1}$ 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 $10^{-29} \text{ cm}^6 \text{ s}^{-1}$ at 150 °K;⁸ a similar value should be appropriate for reaction (3).

The portion of free Cs will then be

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

For illustration, if at 80 km, $n = 5 \times 10^{14} \text{ cm}^{-3}$, $[O_2] = 10^{14} \text{ cm}^{-3}$, $[O] = 10^{11} \text{ cm}^{-3}$, and $k_4 \sim k_5 \sim 3 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$, we have

$$\frac{[Cs]}{[CsO + CsO_2]} \sim 3 ,$$

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 k_4 and k_5 , this qualitative result is compatible with the earlier cesium seeding experiments² 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 O₃ abundance above 80 km of 10¹⁴ molecules/cm², and an absorption cross section of $\sigma(\text{O}_3|3200 \text{ Å}) \approx 3 \times 10^{-20} \text{ cm}^2$, the Cs deposited in this region will not be seriously shielded from sunlight. The Cs photoionization cross section near the threshold⁹ is $\sigma \approx 10^{-19} \text{ cm}^2$. With an average solar flux of 10¹³ photon/cm²-s-Å and a region of the order of 1000 Å wide free of attenuation of O₃, the ionization rate is

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

or an ionization time of about 1/4 hour.

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



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 O⁻, O₂⁻, 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

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

a negative ion ratio of

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

and a cesium ion/neutral ratio of

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

An electron density of $3 \times 10^7 \text{ cm}^{-3}$, 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 $N_e \sim 10^5 \text{ cm}^{-3}$ 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.⁹

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.¹⁰ 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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