## Heptyl aerosolization in regions of fall of separated stages of carrier rockets

A.P. Sadovskii, V.F. Raputa, S.E. Ol'kin, S.V. Zykov, and I.K. Reznikova

State Scientific Center of Virology and Biotechnology "Vektor," Scientific Research Institute of Aerobiology, Novosibirsk Region Institute of Computational Mathematics and Mathematical Geophysics, Siberian Branch of the Russian Academy of Sciences, Novosibirsk

Received February 9, 2000

The regions of fall of separated stages of carrier rockets launched from Baikonur cover an area about 4.5 million hectares at the territory of Russia. Soil, surface water bodies, and flora of the fall regions are inspected for pollution by heptyl rocket fuel. However, heptyl transport in the atmosphere due to aerosolization remains beyond inspection. The comprehensive study of the ecosystem near the fall region FR 213 in the Novosibirsk Region revealed deterioration in the health structure of population, which is supposedly caused by processes of heptyl aerosolization in the atmosphere. Some approaches to studying heptyl aerosolization at fall of separated stages of carrier rockets are proposed.

### Introduction

The regions of fall of separated stages of carrier rockets launched from Russian space-vehicle launching sites have an area of about 20 million hectares. For instance, the fall regions of the Baikonur space-vehicle launching site cover an area of 4.5 million hectares, including 0.96 ha in the Novosibirsk Region, 1.96 ha in the Tomsk Region, and 0.53 ha in the Republic of Altai.

A zone adjacent to the fall regions includes agricultural lands, oilmen's settlements, and reserves. The heptyl component of the rocket fuel, whose basis is non-symmetric dimethylhydrazine (NDMH), presents the main hazard for the fall regions. Separated stages of carrier rockets can contain roughly from 600 to 1300 liter dimethylhydrazine. Thus, the fall regions are regularly subjected to a chemical substance that is not typical for the corresponding ecosystems. The influence of rocket fuel components on environmental objects and human health in the fall regions is studied very poorly. This is explained both by the secrecy of the subject connected with rocket technology and by technical difficulties accompanying observation and collection of necessary information in the fall regions.

Recently, the problem of heptyl pollution of the environment and possible consequences for human health are widely discussed in mass media in connection with conversion of rockets and utilization of rocket fuel. This problem is especially urgent for people living in the regions of fall of separated stages of carrier rockets, since the space activity obviously will not be decreased in the nearest future.

The absence of any systematized information about possible consequences of steady ingress of non-symmetric dimethylhydrazine and its products in the ecosystem does not permit one to justify and formulate the criteria of ecological safety for the fall regions and, all the more, to justify the regional concept of stable development for these areas.

Reference 1 gives a rather full survey of the results of study of possible mechanisms of NDMH influence on the human organism. It is found that NDMH has a toxic effect on liver, nervous system, and immune status of people. Besides, non-symmetric dimethylhydrazine possesses carcinogenic and mutagenic effects. However, the products of NDMH transformation in the atmosphere and water systems are far more dangerous. It is found<sup>1</sup> that non-symmetric dimethylhydrazine (up to 60%) is into transformed dimethylnitrosoamine in the atmosphere. The half-period of transformation strongly depends on the state of the atmosphere, the presence of ozone, hydroxyl radicals, nitrogen dioxide, and the air temperature and varies from several minutes to several hours. The dimethylhydrazine products in water systems are dimethylnitrosoamine, formaldehyde, and dimethylamine. 1 The half-period of transformation in water is tens of days and depends on the presence of metal ions and organic compounds in the water medium and on the water temperature. Among the NDMH products, dimethylnitrosoamine is the most dangerous substance, as it possesses the pronounced carcinogenic and mutagenic effects. Thus, the problem of ecological safety of the fall regions includes at least three aspects:

- 1. Development and improvement of chemicalanalytical methods for detecting NDMH and its products in environmental objects;
- 2. Study of possible ways of transport of NDMH and its products in the environment;
- 3. Ecological-epidemiological studies in the fall regions and revealing of cause-and-effect relations.

In this paper, we attempt to solve this problem although partially. The study was performed in the interests of the military-space forces based on the requirements specification given by specialists of the Department of Geography of the Moscow State University. The Chistoozernyi and Severnyi districts of the Novosibirsk Region were the subjects of the study. The present-day monitoring system of the fall regions is

Optics

based on analyses of heptyl content in the soil, water systems, and plants. The problem of heptyl aerosolization in the lower atmosphere and possible ways of its atmospheric spread were still beyond consideration and not included into the monitoring system.

# Chemical-analytical study of heptyl pollution of environmental objects

According to the existing scheme, environmental objects in the fall region in the Chistoozernyi district of the Novosibirsk Region (FR 213, "Tsyganskii bereg") were studied, and the morbidity of people living near

 $R_1$ 

this region was analyzed. The FR 213 territory is situated in the southern part of West Siberian Plain. The terrain of this region is not uniform. Its northeastern part studied in this paper is a low weakly wavy plain with rare narrow ridges from 3 to 8 m high. The river system is almost absent on the FR territory. A considerable part of the FR is occupied by lakes that are not deep, flat-bottom, and often drying up in summer. Mineralization of the lake water is different: salt and acrid. Due to salinity of soil-forming rocks, soils of the saline type, namely, meadow-chernozem, alkaline chernozem, solonetz soil, and saline soil, are widely occurring in FR 213.

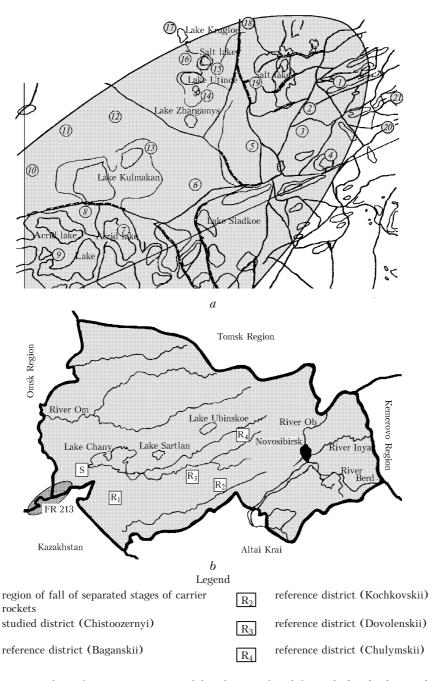


Fig. 1. Arrangement of sampling points in FR 213 (a) and geography of the studied and reference districts (b).

Environmental objects and sampling sites were chosen taking into account our experience of such studies in the Novosibirsk Region and the results of previous investigations of FR 213. We collected 17 samples of surface waters, 21 soil samples, and 21 samples of flora. The geography of sampling is shown in Fig. 1a. Water samples were preserved by hydrochloric acid (at the rate of 5 ml of 33% HCl per 1 liter of water).

Non-symmetric dimethylhydrazine (heptyl) in the collected samples was determined in the form of their pentafluorobenzoylchloride derivatives by the method of gas chromatography with mass-spectrometric detection.

The results of the analyses are presented in Table 1. The heptyl detection limit of the applied method is 0.1  $\mu g/kg,$  and the values of the maximum permissible concentration for heptyl are 10 µg/l in water and 100 µg/kg in soil.

Table 1. Content of 1,1-dimethylhydrazine in samples of surface waters and plant collected in FR 213 "Tsyganskii bereg"

Number of sampling point	NDMH content, μg/l	
in Fig. 1	Water	Plant
1	0.2	0.4
2	< 0.1	< 0.1
3	< 0.1	< 0.1
4	< 0.1	< 0.1
5	< 0.1	< 0.1
6	< 0.1	< 0.1
7	< 0.1	< 0.1
8	0.3	< 0.1
9	-	0.2
10	-	< 0.1
11	-	< 0.1
12	< 0.1	< 0.1
13	0.2	< 0.1
14	< 0.1	< 0.1
15	< 0.1	< 0.1
16	< 0.1	< 0.1
17	< 0.1	0.3
18	0.4	0.9
19	-	< 0.1
20	< 0.1	< 0.1
21	< 0.1	< 0.1

Note. The heptyl content in soil is  $< 0.1 \,\mu g/kg$  in all the samples.

As is seen from Table 1, the measured heptyl concentration was significant at only a few points (marked by a bold face), but it is much less than the maximum permissible concentration.

Thus, the heptyl influence on human health through soil, water, and plants can be excluded with high probability.

### Ecological-epidemiological analysis in the zone adjacent to the fall region

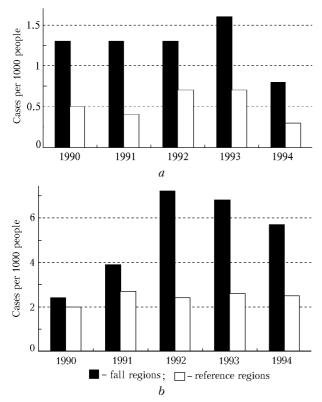
According to the data of civil defense staffs (Russian Ministry of Emergency), about 100 separated stages of carrier rockets launched from Baikonur fell down in the Chistoozernyi district of the Novosibirsk Region during the last 13-15 years. A falling second stage contains from 600 to 1500 kg of residual heptyl. Thus, separated stages brought about 100 t of heptyl in the Chistoozernyi district for the period mentioned above. Taking into account that 1,1dimethylhydrazine rather quickly transforms into dimethylnitrosoamine in the atmosphere, one can suppose that such a great amount of dangerous toxicants in the fall regions must affect the health of peoples living near the region.

The human population is a very important element of the ecosystem and is distinguished by the largest amount of collected information since offices of the public health system regularly record the parameters of human health. Consequences of the effect of the components of rocket fuel and its products on the environment can be followed in the data of ecologicalepidemiological analysis of the human health structure in the corresponding regions. The toxic effect of rocket fuel components on a human organism manifests itself in diseases of the nervous system, liver, kidneys, and blood. However, the highest risk for the health is connected with products of transformation of nonsymmetric dimethylhydrazine into dimethylnitrosoamine, which possesses pronounced carcinogenic and mutagenic effects.

The ecological-epidemiological analysis of the fall regions was performed by the following scheme. In the Novosibirsk Region, we have chosen four reference (Baganskii, Dovolenskii, Chulymskii) that have close climatic and geographic characteristics, similar industrial infrastructure, similar levels of medical service and social parameters with the fall region but are situated rather far from it. Then, using the data of the Novosibirsk Regional Committee on Environmental Protection and Natural Resources and those of the Novosibirsk Regional Committee on Public Health, medical-demographic parameters in the fall region and reference districts were analyzed comparatively. Figure 1b shows the geography of the studied region and reference districts.

Figure 2 shows the results of comparison of two parameters: congenital anomalies in children and neoplasm level. Samples collected for five years were used. As is seen from the histograms, the parameters of the fall regions are significantly worse as compared to the reference districts. This fact is in agreement with the existence of the "heptyl factor" in the fall regions presence can be explained by the and dimethylnitrosoamine, a product of dimethylhydrazine, in the environment. This substance possesses both the carcinogenic and mutagenic effects.

An even more significant difference between the fall regions and the reference districts is observed when comparing the results of correlation analysis for some principle medical and demographic parameters. The analysis was performed by the following scheme.



**Fig. 2.** Results of comparative medical-demographic analysis for the studied and reference districts: level of total morbidity of children (neoplasm) living in the Novosibirsk Region (a); cases of congenital anomalies in children living in the Novosibirsk Region (b).

The dynamics of health parameters during the last 20 years (from 1976 to 1996) was considered for the Novosibirsk Region as a whole. This information was then used as a base for comparison. Then, analogous parameters of the reference districts and districts adjacent to the fall zone for the same period were compared with respect to the comparison base. The results of the correlation analysis are presented in Table 2.

One can see that the dynamics of parameters for the reference regions correlates well with the corresponding parameters for the Novosibirsk Region as a whole (correlation coefficient is 0.8–0.9), and, at the same time, the correlation with the fall regions is low or fully absent. Thus, we can conclude that the presence of the "heptyl factor" in the human environment has a significant effect on the human health and not in a good way.

Comparing the geography of the studied district (Chistoozernyi) and the reference districts (see Fig. 1b) and the data of Table 1, we can conclude that soil, surface waters, and plants can be excluded from the list of possible ways of heptyl spread leading to the negative effect on the human health. As it was mentioned above, the aerosol way of heptyl spread in air remains beyond monitoring.

There is a good reason to suppose that this way affects most strongly the human health. First of all,

this is indicated by the medical and demographic parameters for the studied and reference regions. If we suppose that residual heptyl in tanks is aerosolized along the falling trajectory of separated stages of carrier rockets, then the observed medical and biological phenomena can be understood and explained. There are very few experimental facts related to heptyl aerosolization. This is explained, first of all, by tremendous technical difficulties in the study of this process.

Table 2. Medical-demographic characteristics of fall regions and reference districts of Novosibirsk Region in 1976-1996

	Correlation coefficient $r$	
Medical and demographic parameters	Fall regions  – reference districts	Region as a whole - reference districts
Total morbidity of children		_
for their first year	- 0.2*	0.9**
Morbidity of children because		
of congenital anomalies	0.3*	0.8**
Deceleration of children's	0.4*	0.9**
growth		
Morbidity of women in		
childbirth and recently confined		
women (anomalies of birth	- 0.3*	0.8**
activity)		
Number of first-aid cases	0.2*	0.8**
Diseases of nervous system	0.1*	0.9**
Morbidity with malignant		
tumors	- 0.1*	0.9**
Part of patients with		
neglected malignant tumors	0.2*	0.9**
Death rate in infants	- 0.3*	0.8**
Part of died persons	0.1*	0.8**

<sup>\*</sup> p > 0.05,

It is well-known that constructions of separated stages of carrier rockets are destroyed at heights of about 30 km. It is also known that some amount of heptyl reaches the Earth's surface together with construction fragments. Emission into the atmosphere occurs at the descent trajectory. However, there are no data on the quantity and dynamics of heptyl emission into the atmosphere, mechanism of aerosolization, and size spectrum of particles. The most adequate method for solving this problem is mathematical simulation with joint consideration of some experimental facts.

# Possible approaches in simulating heptyl aerosolization

In this section, we outline the main approaches to solution of the problem of aerosol spread from high-altitude sources. Depending on the input information, both direct and inverse problems of aerosol transport in the atmosphere can be formulated. For direct simulation, the following parameters of the source must be described correctly: aerosol emission, position of the

<sup>\*\*</sup> p < 0.05, confidence of correlations.

source in space and time, physical and chemical properties of the aerosol admixture. Besides, hydrological and meteorological conditions must be known. Depending on the purposes, the following ways of setting meteorological conditions are possible here:

- 1. Description of wind velocity, temperature, pressure, and air density on the basis of the dynamic equations of the mesosphere, stratosphere, and troposphere  $^{2,3}$ ;
- 2. Determination of actual statistic characteristics of the atmosphere in the layer of spread of aerosol particles  $^{4-6}$ :
- 3. Use of current values of principal meteorological parameters obtained from direct observations.

The third way is preferable as it allows the highest accuracy in determining aerosol pollution fields to be obtained. However, its implementation meets severe difficulties. The first two ways of setting the meteorological conditions are convenient for use when solving scenario problems of aerosol pollution of an area subject to high-altitude emissions. In direct simulation of rocket fuel transport in the atmosphere, one should not expect high accuracy because of large uncertainty in position of the emission source, its operating conditions, and aerosolization of heptyl in the atmosphere.

In our opinion, a complex approach involving both experimental studies of heptyl pollution of territories and theoretical concepts of aerosol spread from high-altitude sources, as well as additional *a priori* qualitative and quantitative information on possible pollution zones is most realistic. It is convenient to describe this approach by formulating the corresponding inverse problems.  $^{7,8}$ 

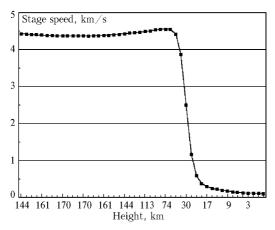


Fig. 3. Speed of the second stage of the rocket vs. height during the passive flight.

Let us consider possible variants of heptyl aerosolization in the atmosphere using as an example the motion of the second stage of the Proton carrier rocket. The motion of the second stage includes active and passive parts of the flight. The active flight lasts about 3.5 min. During this time, the rocket moves from

the height of 44 up to 144 km and acquires the speed of  $4.4~\rm km/s$ . Then, at this height, the second stage is separated, and the passive flight begins. The fuel remaining in tanks can weight more than half-ton. Heptyl aerosolization is more probable during the passive part of the flight.

Figure 3 shows how the speed changes with height during the passive flight of the second stage.

As is seen from Fig. 3, the stage moves along the ballistic trajectory in the upper layers of the atmosphere. When it enters denser atmospheric layers, its speed decreases due to friction, at the height of 25–30 km the speed decreases sharply, and fuel and oxidizer tanks explode. This can lead to stage destruction and intense heptyl emission at these and lower heights. Further atmospheric transport of heptyl depends significantly on the results of its aerosolization. We should note two extreme cases corresponding to the following sedimentation rates<sup>9</sup>:

(a) 
$$U_{\rm m}/W \leq 10$$
, (b)  $U_{\rm m}/W > 60$ ,

where  $U_{\rm m}$  is mean wind speed, and W is sedimentation rate. Correspondingly, kinematical and diffusion spread schemes take place.

In the case (a), the maximum heptyl emission should be expected at the distance  $X_{\rm max}$  in the direction of the mean wind  $U_{\rm m}$  from the place of tank explosion. The value of  $X_{\rm max}$  is estimated by the following relation<sup>9</sup>:

$$X_{\text{max}} \leq H U_{\text{m}} / W$$
,

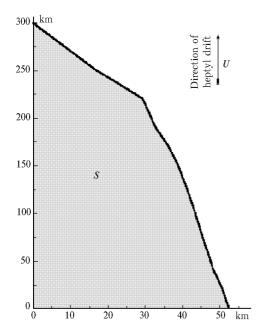
where H is emission height. If we suppose that  $U_{\rm m}/W \le 10$ , H=30 km, then the position of  $X_{\rm max}$  can vary within 300 km.

In the case (b), processes of vertical and horizontal turbulent diffusion have a significant effect on the spread of an admixture. Heptyl fallout should be expected on a very large territory, and  $X_{\rm max}$  can be more than 1000 km (Ref. 10). The density of aerosol fallout is very low in this case and varies weakly within the territory.

Figure 4 shows the zone S of possible kinematical fallout of rocket fuel  $(U_{\rm m}/W \le 10)$  in the case of continuous heptyl outflow. The horizontal axis is a projection of the stage fall trajectory onto the Earth's surface. The origin of the coordinate system corresponds to the point on the Earth's surface, above which the tanks exploded. The point of intersection of the curve outlining the area S with the horizontal axis corresponds to the place of stage fall. The direction of the vertical axis coincides with the vector of mean wind velocity in the layer of heptyl fallout.

Since the rate W in the upper atmosphere considerably exceeds that in dense atmospheric layers, the area of possible heptyl fallout shown in Fig. 4 is an upper bound. Closer mathematical consideration of the conditions of heptyl sedimentation and dispersion can give a more accurate estimate of the fallout area. It

should be noted that the best conditions for experimental study of pollution of a territory are in the case when the direction of the mean wind and the stage fall direction are close. In this case, the area of possible pollution can be minimal.



**Fig. 4.** Area *S* of possible kinematical heptyl fallout.

In conclusion, we would like to emphasize the following statement. On the one hand, the results of analysis of soil, water, and flora show that the negative effects on the ecosystem are minimal. On the other hand, marked changes are observed in the structure of health of people living in the regions adjacent to fall zones. So, we can conclude that the problem of heptyl aerosolization in the fall zones and adjacent regions comes to a leading place in the system of monitoring of fall zones.

#### References

- 1. G. Choudhary and H. Hansen, Chemosphere **37**, No. 5, 801–843 (1998).
- 2. J.R. Holton, *The Dynamic Meteorology of the Stratosphere and Mesosphere* (American Meteorological Society, 1975).
- 3. D.L. Laikhman, ed., *Dynamic Meteorology* (Gidrometeoizdat, Leningrad, 1976), 608 pp.
- 4. E.P. Shkol'nyi and L.A. Maiboroda, *The Atmosphere and Control of Aircraft Motion* (Gidrometeoizdat, Leningrad, 1973), 307 pp.
- 5. B.N. Charles, J. Meteorol. 16, No. 5, 581-593 (1959).
- 6. L.V. Kudryavtseva, Meteorol. Gidrol., No. 3, 41-50 (1982).
- 7. A.I. Krylova, V.F. Raputa, and I.A. Sutorikhin, Meteorol. Gidrol., No. 5, 5–13 (1993).
- 8. V.F. Raputa and A.I. Krylova, Meteorol. Gidrol., No. 2, 49–58 (1995).
- 9. G.M. Petrova and A.N. Miroshkina, Trudy IPG, No. 7, 5-40 (1967).
- 10. Yu.A. Izrael', A.S. Volkov, and A.F. Kovalev, Meteorol. Gidrol., No. 5, 5–11 (1995).