Experimental study and numerical modeling of fluorine distribution in Bratsk region

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Fluorine concentration fields in the vicinity of Bratsk Aluminum Plant were calculated with the use of mathematical model. Calculation and modeling results are compared. The intensity of fluorine sink on the underlying surface of the Bratsk region is estimated.

Bratsk Aluminum Plant (BAP) has been functioning since 1966 using the technology of self-calcinating anodes (Soderberg technology) and wet gas purification system. Final gases consist of polyaromatic carbohydrates, perfluorocarbons, CO_2, SO_2, HF, SiF_4, solid fluorides (cryolite, calcium fluoride, aluminum fluoride), and other components. The major part of fluorine is caught in wet gas purification system and returns to the production process as secondary cryolite, while the rest of fluorine is emitted in the environment.[1]

Modern technology uses self-calcinating anodes and dry gas purification system in order to reduce the emission. Average recommended production quantity in foreign countries is limited to 200–300 thousands of tons a year.[2] In 2005 yearly aluminum production at BAP was approximately 975 thousands of tons.

Excess of fluorine in human body leads to bone disease fluorosis; its deficiency causes caries. The authors of Ref. 3 revealed that the level of musculosceletal system diseases of population in Bratsk is by 43.8% higher than in Irkutsk Region. Of dangerous are biologically active mobile forms of fluorine capable of intruding into trophic chain accumulating in plants.[4] That is why the study of spatiotemporal variations of gas fluoride concentration (in terms of fluorine) and the value of its sink on the underlying surface is very urgent.

Bratsk is situated in the southern part of the Middle Siberian plateau, consisting of separate residential areas situated on both banks of the Bratsk water reservoir (Fig. 1).

The climate is sharply continental. The ground is under stable snow cover from October to March having 150–180 days bedding period.[5] The hilly relief has height differences from 400 to 670 m. The Dolgii mounting range is situated to the south and south-west from the plant. Dominating wind direction is western and south-western with a wind speed of 1–5 m/sec. Thus, the plant emissions are mostly directed to residential areas.

Fig. 1. The location of towns and population areas in the vicinity of Bratsk.

The transport of different aerosol fractions over Bratsk and the calculation of the hazard city pollution function were earlier studied in Ref. 6, where the results of distribution and sink of fluorides from the Bratsk Aluminum Plant were investigated using measurements[7,8] and mathematical model calculations.[9] The data on anhydrous hydrogen fluoride concentrations in atmospheric air, as well as the data on wind direction and speed were taken from Refs. 5 and 7.

The fluorine emission distribution was modeled using a special-purpose numerical model. We selected an area of 60×40 km^2 and of 2400 m altitude over the surface of the Bratsk water reservoir. The following parameters were used in calculations: the time and horizontal steps were 5 min and 2 km, respectively; vertical step was equal to 50 m up to an altitude of 400 m from the reservoir surface, then 100 m up to 600 m; and further it was 300, 500, and 1000 m.

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The first series of numerical experiments was aimed at studying the influence of the wind speed and direction on fluorides distribution within the investigated area. The intensity of fluorine emission was determined by the material balance method. It was revealed that the Dolgii mountain range, located to the south and south-east of the source influences distribution of the fluoride emissions. In case of the winter wind the concentration of fluorides along the flow is less at the same distance from the source than in the case of western and west-north-western winds. Note that wind flow of northern and eastern directions is observed more often in autumn-summer period.

Figure 2 illustrates calculated averaged surface concentrations of anhydrous hydrogen fluoride in the investigated region.

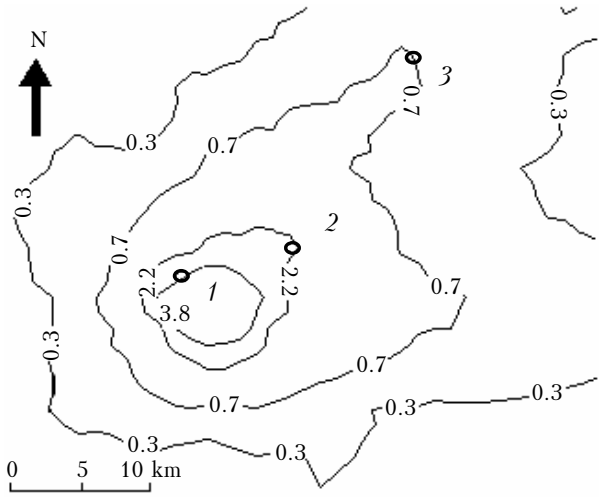

Fig. 2. Isolines of calculated averaged surface concentrations of anhydrous hydrogen fluoride in Bratsk region, μg/m^3: Chekanovskii settlement (*1*); Telecenter (*2*); Padun (*3*).

The variations in wind speed for 16 gradations were taken into account. In order to validate the results of numeric modeling, the calculated values of anhydrous hydrogen fluoride concentrations were compared with the data of BCHMS (Bratsk Central Hydrometeoservice) on HF concentrations in air of different regions of Bratsk, averaged over 2002 (Table 1).

Table 1. The comparison of experimental and calculated values for fluorine concentrations in air

The number of sampling site	Fluorine concentration values, μg/m^3	
	measured	calculated
1	3.8	3.8
2	1.9	2.2
3	0.95	0.7

The comparison of anhydrous hydrogen fluoride values, calculated and measured at BCHMS posts, revealed a satisfactory correlation.

The second series of numerical experiments was conducted for determination of the fluoride sink intensity on the underlying surface of the Bratsk region. The fluorine sink rate was estimated using the data on anhydrous hydrogen fluoride content in air and the data on the intensity of water-soluble fluorine sink on the investigated territory. It was found that the value of fluorine sink rate is between 0.1 and 0.6 cm/sec. In calculations we took the value of fluorine sink rate to 0.2 cm/sec. The repetition of different wind gradations by directions was also taken into account. Figure 3 illustrates calculated values of fluorine sink intensity in Bratsk region measured in kg /(km^2 · month).

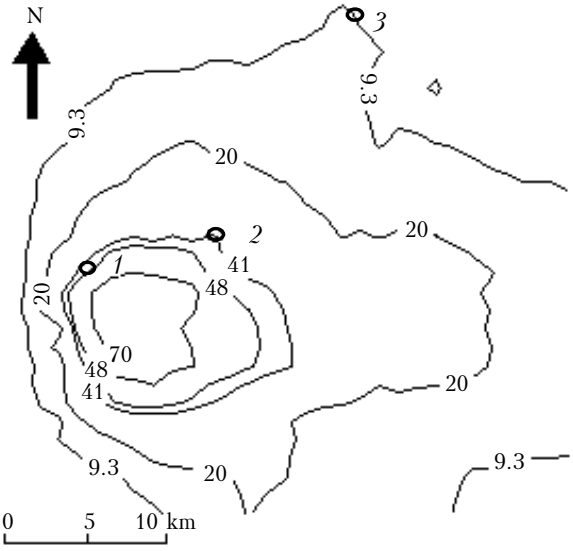

Fig. 3. Isolines of calculated values of fluorine sink intensity in Bratsk region measured in kg/(km^2 · month).

Fluorine sink intensity higher than 40 kg/(km^2 · month) was registered in BAP region on the area of 125 km^2, and higher than 70 kg/(km^2 · month) — on the area of 35 km^2. To validate the estimation of soluble fluorine sink intensity by the model, the obtained values were compared with annual average values of global observation data for the period 2000–2005 [Ref. 8], (Table 2). The comparison has shown a satisfactory correlation.

Table 2. The comparison of measured and calculated values of soluble fluorine sink intensity at the territory of Bratsk region in winter period

The number of sampling site	Fluorine sink intensity, kg/(km^2 · month)	
	Measurements data	Model calculation
1	58	48
2	50	41
3	15	9.3

Based on the results of calculations and measurement data, the values of fluorine mass sank in the investigated region were estimated. It was found, that about 17% of the emitted fluorine was sunk on 60×40 km^2 area in the Bratsk Aluminum Plant region for a year. Approximately, one third of this fluorine is trapped by ground undulations. In case of plain surface, the amount of fluorine emitted in the atmosphere by BAP and sunk on this area would be 12%. Note that according to the results of snow-chemical survey conducted in the region of Novokuznetsk and Bratsk Aluminum Plants, about 15% of the total amount of fluorine emitted in the atmosphere is sunk in a radius of 15 km.[10]

Thus, the investigation conducted has broadened our knowledge of the conditions of distribution and sink in Bratsk region of fluorine compounds. Quantitative characteristics of fluorine sink on the underlying surface of the investigated area were estimated. The comparison of measured and calculated with a mathematical model values of anhydrous hydrogen fluoride concentrations in atmospheric air with soluble fluorine sink intensity values revealed their satisfactory agreement. It has been also found that the local relief impedes the transport of emissions to the south and to the west from the plant.

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