Annals of Agrarian Science

Journal homepage: http://journals.org.ge/index.php

Determination of the ratio hematite/goethite by soil color

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Received: 10 September 2018; accepted: 12 November 2018

A B S T R A C T

The content and composition of hematite and goethite can solve important tasks of genesis, as well as soil classification. It is proposed a new method of determining the index I_{Lab} = [Hematite/(Hematite+Goethite)] on color soil system CIE-L*a*b*. The method is based on calculating soils Redness (Red) and then calculating the I_{Lab}, based on reference Red of hematite and goethite. Replacement of Fe^{3+} to Al^{3+} is taken into account through amendments to the color of goethite, admitting that Al-hematite color does not differ from color pure hematite. The new technique was proven on Northern Italy andosols and was give results similar to results, received for converting optical spectrum on Kubelka-Munk theory. But the new technique has the advantage of ease of calculation and the ability to use the old color data obtained in the Munsell system. Validating new methods in luvisols is showed an agreement index values I_{Lab} with real [Hematite/(Hematite+Goethite)] in those parts where there are profile have pure hematite and goethite. In luvisols containing Al-goethite and Al-hematite, admixture Al generates an error in the calculation. To delete the error, it is proposed an amendment to the distorting effects of aluminum in lattice goethite, ewith high content: 8-12 mole Al %. Cambisols have a value index I_{Lab} also vary depending on the impurities Al in hematite and goethite. From samples with particles of pure goethite Red higher than cambisols containing Al-goethite. After adjustment for the Al-share in goethite the share of hematite becomes comparable to its share in the cambisols samples with pure goethite.

Keywords: Hematite, Goethite, Luvisoils, Cambisols, Genesis, Geochemistry

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Introduction

Hematite αFe_2O_3 and goethite αFeOOH as the most stable Fe-(hydr)oxides forms, are widely distributed in soils of different genesis and age. To the greatest extent Fe-(hydr)oxides are concentrated in the oxisols and cambisols [1-3], and also – in many loess soils [4, 5]. Content of hematite and goethite is useful to solve important tasks of genesis and classification: for example, to distinguish between oxisols and cambisols [1].

At the same time, genesis and geochemistry of these αFe-minerals are varies considerably. So, goethite is formed in acidic environment and in conditions of high humidity and moderate temperature, whereas hematite is formed in neutral conditions and with low humidity and high temperature [6]. This opens up opportunities to the ratio hematite/goethite to determine a soil evolution history. Huge significance for geochemistry has Fe substitution for Al in the lattice of these minerals, particular for goethite. For example, the degree of substitution of aluminum associated with age of alluvial soils (Victoria, Australia) [7]. In the more ancient soils with age 760 thousand years Al content was higher (8-12 mol %) than in young soils with age ~ 40 thousand years, where the Al contents was only 3.5-5 mol %. There are other important differences in the content of aluminum in soils with goethite of different genesis. Comparison of the proportion of Al in goethite of alfesols (Indiana, United States) and oxisols (Goya, Brazil) showed a statistically significant difference. Oxisols goethite is contain 11-14 mole Al %, whereas alfisols goethite – only 7-9 mole Al % [8].

Accurate information on soil goethite and hematite gives Mossbauer spectroscopy, especially in a strong cooling, to the temperature of liquid nitrogen – 4 k [6, 9]. In addition to the percentage of Fe in goethite and hematite Mossbauer spectroscopy is allows you to judge the degree of particles mess structure, that is associated with the destructive influence of Al or Ti impurities on the (hydr)oxides structure. With the development of spectrophotometers it is appeared able to exact number-definition of soil color and judge the correlation of main soil pigments. These include: humus as a black pigment, carbonates as a white pigment, Fe-(hydr)oxides as red and yellow pigments [10]. It is primarily red hematite αFe_2O_3 and yellow goethite αFeOOH. It is asked a few techniques using optical spectrum of soil to determine the hematite and goethite share. The end result of the analysis is usually expressed as the ratio [Hematite/(Hematite+Goethite)]. Let us name this ratio as index I.

Optical-mineralogical methods of an index I determination is based on the ratio of red and yellow parts of the soil spectrum. The most common technique is based on converting optical soil spectrum using equations of Kubelka-Munk [11, 12, 13]. Then a new spectrum is differentiated twice and compares the amplitudes of two reflexes: 530 nm for hematite and 413 nm for goethite. Attitude [Hematite/(Hematite+Goethite)], calculated by this method are denoted by index $I_{K\text{-}M}$. This method can serve as a benchmark in determining the ratio of hematite and goethite on optical spectra of soil. Disadvantage: the complicated procedure is requirement to use a computer programs.

At present, the color of the soil is characterized mainly by two optical systems: earlier Munsell system and more modern system of CIE-L*a*b* [10, 14-16]. Munsell color point is defined by three characteristics: Hue (H), Value (V) and Chroma (C). Because no equivalent perception of light and dark color tones all cylindrical color space is filled.

In the CIE-L*a*b* system: L* is coordinate set of lightness (varies from 0 to 100, from the darkest to the lightest), chromatic component is defined by two Cartesian coordinates a* and b*. The first coordinate is indicated the position of the colors range from green (-a*) to the red (+a*), and the second is ranged from blue (-b*) to yellow (+b*) [3, 14]. Coordinates a* and b* are distinguished clearly contribution of Fe-minerals to the soil color.

Since orthogonal CIE-L*a*b* system is much better then convenient cylindrical Munsell system [3, 10, 14], then the color of minerals and soils we express only in CIE-L*a*b* system. Data sources, expressed in Munsell, we were transferred to CIE-L*a*b* system, algorithms re-accounts is published in papers [17, 18]. Soil scientists have gathered information about the Munsell color of the soil, published in numerous articles. But learn from them hematite and goethite information using known approaches is impossible. Therefore, we are proposed a new approach that would use previously received information about Munsell color to extract information on the ratio in soils of hematite and goethite. Of course, the new technique is suitable for counting relations [Hematite/ (Hematite+Goethite)] in soil samples, with modern spectrophotometer issuing the result immediately in the CIE-L*a*b*. Ratio [Hematite/(Hematite+Goethite)] is calculated on new methods, let the index of I_{Lab}.

The objective of this study: to propose a new method for determining the ratio hematite/goethite by the soil color in the CIE-L*a*b* system.

Objects

We are analyzed Fe-(hydr)oxides using dashboards Sheinost and Schwertmann [19] on reference samples (hydr)oxides of iron (n = 277). Among them: hematite αFe_2O_3, maghemite γFe_2O_3, goethite αFeOOH, lepidocrocite γFeOOH, ferrihydrite $2Fe_2O_3 \cdot FeOOH \cdot 4H_2O$ and feroxyhite δFeOOH. Their colorimetric characteristics in the source were expressed in Munsell, we counted them in CIE-Lab system.

To compare two methods: 1) with the conversion of optical spectrum according to the Kubelka-Munk functions, and 2) new techniques we were used the color data of modern andosols and paleosols in Northern Italy [20].

Basic researches are devoted to the analysis of luvisols of southern Spain, the source data is published in [2]. In addition, researches are devoted our data on color of cambisols from Lithuania, and Arkhangelsk, Perm and Vologda regions (Russia).

Methods

The methodology of calculating the ratio of hematite/goethite by the soil color in the CIE-Lab System. First of all, the minerals color it is necessary to express one number.

As can be seen from the table 1, Fe-minerals is shown redness (a*) and yellowness (b*), although in different proportions. The ratio between the redness (a*) and yellowness (b*) use as an indicator (Red) of each mineral and soil sample; let us express Red it in the form:

Red = [a*/(a* + b*] (1)

Value (hydr)oxide Red is reduced in this order: hematite ($αFe_2O_3$) > feroxyhite (δFeOOH) > ferrihydrite > (2Fe2O3·FeOOH·4H2O) > maghemite ($γFe_2O_3$) > lepidocrocite (γFeOOH) > goethite (αFeOOH). Red of hematite is maximal: Red = 0.51, and Red of goethite is: Red = 0.16. The degree of Red of hematite have expressed about 3 times stronger than the goethite. We emphasize that these values Red was obtained on a pure Fe-minerals.

Content of hematite and goethite was studied in soils enriched Fe-(hydr)oxides: oxisols, cambisols, etc. [2, 6]. Their ratio is usually expressed in the form: [Hematite/(Hematite+Goethite)].

This relationship you can count on soil color CIE-Lab system using index Red. In soils with two contrasting pigments: red hematite and yellow goethite Red index is range from 0.51 in the presence of single hematite to 0.16 - in the presence of single goethite. Intermediate index values Red will have samples with different fractions of hematite and goethite in the absence of other Fe-pigments in the soil. The second condition: minimum index value Red = 0.16 meets soils with pure goethite, but if there is only one Al-goethite Red value falls below 0.16.

Attitude [Hematite/(Hematite+Goethite)] defined by the new methodology is based on soil color CIE-Lab system will be denoted by I_{Lab}. The index of the I_{Lab} races-read from the formula:

I_{Lab} = 1 – [(0.51 – Red)]/(0.51 – 0.16), (2)

where is 0.51 – averaged magnitude of hematite Red and 0.16 – averaged magnitude of goethite Red.

In soils, especially tropical ones, occur Fe-(hydr)oxides with a partial substitution of Fe^{3+} by Al^{3+}. Aluminum is reducing hue of ferruginous minerals. Amendments to the color of both minerals are difficult. Let us confine adjusted only one mineral, where the substitution of iron as much as possible, i.e. to take into account the effect of Al on the goethite color. We are assume that the color of Al-hematite is not different from the color of pure hematite (due to the low degree of substitution of iron in hematite), and for distorting effect on the color of the aluminum is meet Al-goethite.

Comparing the new technique with the methodology of calculating the ratio of hematite/goethite by Kubelka-Munk equation. For this comparison, we are used work [20], where counting relations [Hematite/(Hematite+Goethite)] based on spectral color information of modern soils and paleosoils (North Italy) using Kubelka-Munk function, as well as Munsell Color. We were changed the color in the CIE-Lab system, and then calculated the index of I_{Lab}. The results of the comparison of two methods of counting relations [Heme-Titus/(Hematite+Goethite)] are given in the table 2.

As can be seen, the mean values of relationship [Hematite/(Hematite+Goethite)] for n = 10 are differ not substantially: I_{K-M} = 0.05 ± 0.01 and I_{Lab} = 0.06 ± 0.02; difference is not authentically P 0.95. The correlation coefficient between the indices I_{K-M} and I_{Lab} is r = 0785.

Thus, the new technique is give results similar to results, received for converting optical spectrum on Kubelka-Munk theory, but has the advantage of ease of calculation and the ability to use the old color data obtained in the Munsell system.

Now let us consider the application of the new methodology for calculating the relationship [Hematite/(Hematite+Goethite)] in soils with pure Fe-minerals and mixed with aluminum for example luvisols and kambisols.

Table 1. *Fe-(hydr)oxides color, expressed in Munsell system and CIE-L*a*b* system. The original data in the paper [19]*

Минерал	Munsell	CIE-L*a*b*			
		L*	a*	b*	Red
Hematite	1.2 YR 3.6/5.2	36.6	20.9	20.4	0.51
Feroxyhite	4.2 YR 3.8/6.0	38.7	19.1	29.3	0.39
Ferrihydrite	6.6 YR 4.9/6.3	50.0	15.4	34.9	0.31
Maghemite	8.3 YR 3.1/3.2	31.4	7.4	18.0	0.29
Lepidocrocite	6.8 YR 5.5/8.2	56.1	18.5	46.4	0.28
Goethite	0.4 Y 6.0/6.9	61.0	8.2	44.1	0.16

Table 2. *Mansell color and CIE-L*a*b* system color of recent andosols (RS) and paleosols (I-IV) in Northern Italy. The values I = [Hematite/(Hematite+Goethite)], calculated based on the Kubelka-Munk equations (I_{K-M}) and based on CIE-L*a*b* system. The original data is in the paper [20].*

Sample (depth, cm)	Munsell	CIE-L*a*b*				I_{Lab}	I_{K-M}
		L*	a*	b*	Red_{Lab}		
RS-A (0-20)	9.6 YR 5.5/3.6	56.7	6.8	25.7	0.21	0.14	0.07
RS-Bw1 (20-40)	9.6 YR 5.4/3.5	56.7	4.6	18.7	0.20	0.11	0.10
RS-C1 (80-100)	9.3 YR 7.0/3.7	71.6	5.8	26.1	0.18	0.06	0.09
I-1 (280-300)	8.9 YR 5.6/4.0	56.7	6.8	24.4	0.22	0.17	0.08
II-1 (460-480)	0.3 Y 6.9/3.8	71.6	4.5	27.1	0.14	0.00	0.00
III-1 (840-860)	9.5 YR 6.5/4.0	66.6	5.4	25.1	0.18	0.06	0.04
III-3 (92-940)	9.8 YR 6.8/4.0	71.6	4.5	25.9	0.15	0.00	0.02
IV-1 (1000-1020)	0.1 Y 6.7/4.1	66.7	4.0	26.1	0.13	0.00	0.01
IV-3 (1040-1060)	9.6 YR 6.6/3.8	66.7	5.4	25.1	0.18	0.05	0.03
IV-4 (1060-1080)	9.4 YR 6.3/3.9	66.7	5.4	25.1	0.18	0.05	0.06
Average						0.06± 0.02	0.05± 0.01

Results and discussion

Relationship between Hematite and Goethite in luvisols

The main feature of luvisols is the texture profile differentiation; clay accumulates in the horizon argic, its color ranges from brown to red, depending on the composition of the Fe-(hydr). Luvisols are holded 500-600 million HA in the world, mainly in (sub)tropical regions [21]. We were analyzed two red-brown color luvisols (profiles RB and MO) in the province of Cordoba in southern Spain, described in [2]. Iron minerals are presented in two color: red hematite and (Al)-hematite and yellow goethite and (Al)-goethite. In the upper part of the both profiles in Fe-minerals part of Fe^{3+} is replaced by Al^{3+}. Replacing is extent lesser (to 7 mole Al % in the iron hematite) and twice stronger (up to 14 mole Al % replaces in goethite). It is obvious that the optical effect of the Al presence in the lattice of goethite is higher than from its iron replacement in hematite. The minerals iron impurities are not present at the profile bottom.

Let us compare the calculated value index I_{Lab} with real attitude [Hematite/ (Hematite+Goethite)]. As can be seen from table 3, at the profiles bottom where there are pure hematite and goethite, almost for all samples I_{Lab} ≈ [Hematite/(Hematite +Goethite)]. This means that the optical properties of goethite and hematite in luvisols are close to standard minerals on Sheinost and Schwertmann [19]. A notable deviation is observed only sample only MO-6 where I_{Lab} = 0.23, whereas [Hematite/ (Hematite +Goethite)] = 0.00. Empire share hematite (when calculating by color), obviously, is due to the increased redness real Fe-minerals in the sample MO-6 compared with reference minerals.

Table 3. *Color and Fe-minerals in luvisols of southern Spain. The original data is in the paper [2].*

Sample (depth, cm)	Al-goethite, %	Al-hematite, %	I	Mansell	L*	a*	b*	Red	I_{Lab}	ΔRed
Profile RB										
RB-11(22-45)	1.65	2.80	0.63	7.5 YR 6/4	61.1	8.4	22.3	0.27	0.33	0.11
RB-12 (45-70)	2.25	3.10	0.58	8 YR 7/4	70.9	7.2	23.2	0.24	0.23	0.12
RB-13 (70-105)	2.80	2.80	0.50	7.5 YR 7/4	70.9	7.9	22.6	0.26	0.28	0.07
RB-14(105-130)	2.20	3.75	0.63	8 YR 7/5	70.9	9.0	29.0	0.24	0.23	0.14
RB-15*(130-155)	4.90	1.65	0.25	8 YR 7/5	70.9	9.0	29.0	0.24	0.23	
RB-16 (155-170)*	4.95	0.90	0.15	9 YR 8/3	80.5	3.8	18.6	0.17	0.03	
RB-17(170-190)*	4.35	-	0.00	1 Y 8/2	80.5	0.8	14.0	0.05	0.00	
RB-18(190-230)*	3.60	-	0.00	2 Y 8/1.5	80.5	0.0	10.9	0.00	0.00	
RB-19(230-260)*	4.40	-	0.00	2 Y 8/2	80.5	0.0	14.6	0.00	0.00	
RB-20(260-300)*	4.15	-	0.00	10 YR 8/2	80.5	1.6	13.4	0.11	0.00	
Profile MO										
MO-1 (0-8)	1.95	3.95	0.67	6 YR 6/5	61.0	12.2	26.3	0.32	0.46	0.07
MO-2 (8-50)	2.10	4.10	0.66	6 YR 7/6	70.9	13.8	32.0	0.30	0.40	0.09
MO-3 (50-65)	1.50	4.80	0.76	5 YR 6/6	61.0	15.8	30.6	0.34	0.52	0.09
MO-4 (65-75)	1.50	3.20	0.68	6 YR 7/5	70.9	11.6	26.4	0.31	0.43	0.09
MO-5 (75-100)	1.40	3.00	0.68	7 YR 7/4	70.9	8.4	22.1	0.27	0.31	0.13
MO-6 (100-145)*	3.10	-	0.00	7.5 YR 7/3	70.9	6.0	16.9	0.26	0.23	
MO-7(145-180)*	2.80	-	0.00	10 YR 8/2	80.5	1.6	13.4	0.11	0.00	

* – goethite and hematite without Al.

Characteristically, that luvisols without hematite in the lower parts of the two rips have a low Red = 0.00-0.26. The higher the goethite, the lower the redness: correlation coefficient dependency on Red from the contents of goethite is negative: r =-0. 58. Thus, in the lower layers, where the only Fe-pigment is goethite, increasing its number is lead to lower the already low Red soil.

At the luvisols top are contained hematite and goethite, both enriched with aluminum. This change of the chemical composition of Fe-minerals is lead to heavy distortion of the results of calculation of I_{Lab} index. In fact, in the samples profile RB share Al-hematite from its amount with Al-goethite is reaches 50-63 % while the index calculation based on sheer colors I_{LAB} goethite is show the proportion of hematite only 23-33%. It is clear that the error is obliged to impurities Al in Fe-minerals because the calculation is based on optical reference analysis, pure particles of hematite and goethite. To correct a mistake will make an amendment to the distorting effects of aluminum on the color of goethite with 8-12 Mole Al %. To a first approximation, the influence of aluminum on the color of hematite (2-5 mole Al %) negligible, let us limited to the influence of Al on the color of the goethite only.

Let us calculate the value soil reduction ΔRed due to the influence of Al on the color of goethite, proceeding from the real relationship between Al-hematite and Al-goethite. To do this, we are transform the equation (2) and make it real relation I = [Hematite/(Hematite+Goethite)]. Reduction of soil ΔRed will be calculated from the equation:

$$\Delta Red = 0.51 - 0.35x\{1 - [Hematite/(Hematite+Goethite)]\} - Red \quad (4)$$

The value of ΔRed for luvisols with Al-goethite are listed in table 3. To RB profile ΔRed values ranging from 0.07 to 0.14, on average ΔRed = 0.11, to MO profile ΔRed values ranging from 0.07 to 0.13, average ΔRed = 0.09. Generalized average ΔRed is 0.10. Thus, Al in goethite is understates the luvisols Red on average 0.10. This ΔRed can be used for correction of Al influence on goethite color in soils containing goethite with 8-12 mole Al %.

The ratio hematite/goethite in cambisols

Characteristics of cambisols are heavy texture and a reddish tone, with a low content of iron compounds [21]. Since usually cambisols is weathered weakly they often receive from legacy particles of hematite and goethite. Cambisols are took 1.5 billion HA in the world, they are the most common soils in the world [21].

We were studied four carbonate cambisols in European Russia: 1) in Pinega district, Arkhangelsk, 2) in Perm district, 3) in Zarasai district, Lithuania, 4) in Cherepovets district, Vologda. The results are shown in table 4. As can be seen, the index value of Red is vary in a wide range: from 0.18 to 0.40. Even wider range index values I_{Lab}: from 0.07 to 0.69. Such a large spread of optical indicators is indicated heterogeneity sampling.

A possible reason is the difference in the chemical composition of goethite in cambisols. Indeed, eight samples have pure goethite and four samples is composed goethite with part of Fe^{3+} substituted for Al^{3+}, these two groups are varying in their optical properties. As can be seen from the table 4, cambisols with pure goethite have high Red = 0.33 ± 0.02, and the I_{Lab} index = 0.50 ± 0.05, that is 50% of hematite. The indicators of soil containing Al-goethite significantly below: Red = 0.22 ± 0.02, and the proportion of 17% total hematite: I_{Lab} = 0.17 ± 0.06. Difference of both soil groups is authentically P 95.

Make amendments in color of cambisols with Al-goethite. To do this we will use the data about the average reduction Red previously received for luvisols: ΔRed = 0.10. If ΔRed = 0.10 to add the cambisols containing Al-goethite, the index value will increase significantly I_{Lab} (table 4). As a result, the average value of the I_{Lab} team cambisols with Al- goethite will increased: $I_{Lab(Corrected)}$ = 0.45 ± 0.06. Now the $I_{Lab(Corrected)}$ index of two samples is matched up and their difference becomes unreliable at P 95.

Thus, the amendment on Al in goethite gives more similar picture: 45% of hematite in the same series as that designs cambisols with pure flocking (50%).

Conclusion

Index I = [Hematite/(Hematite+Goethite)] is widely used in the study of the genesis and soil classification. In addition to the mineralogical it is known the optical method of obtaining I index using Kubelka-Munch equation. We are proposed a simpler method of determining I_{Lab} index on soil color using the optical CIE-L*a*b* system. The method is based on calculating soils Redness (Red) and then calculating the I_{Lab}, based on reference Red

Table 4. *Color and Fe-minerals in the cambisols of the Russian plain. The original data is in the book [10].*

Horizon (depth, cm)	Fe-minerals	Fe_{Tot} %	Fe_{DCB} %	L*	a*	b*	Red	I_{Lab}	$I_{Lab-corr}$
Pinega district, Arkhangelsk, Russia									
BM (7-16)	Hematite, goethite	2.41	1.29	60.3	14.5	26.3	0.35	0.55	
B (16-24)	Hematite, goethite	3.65	2.22	57.8	16.8	25.2	0.40	0.69	
D_{Ca} (44-62)	Hematite, goethite	1.00	0.48	71.9	12.0	30.4	0.28	0.35	
Perm district, Russia									
AY (7-12)	Hematite, goethite	4.18	1.57	58.9	11.1	24.7	0.31	0.43	
B1 (19-36)	Hematite, goethite	5.29	2.00	57.6	15.4	25.8	0.37	0.61	
B2 (36-56)	Hematite, goethite	5.34	2.20	56.6	15.8	24.9	0.39	0.65	
Zarasai district, Lithuania									
PYg (0-10)	Hematite, Al-goethite		1.42	66.2	8.0	29.3	0.21	0.16	0.43
$B1_{Ca}$ (40-50)	Hematite, goethite		1.09	67.9	11.3	29.0	0.28	0.35	
BC_{Ca} (80-100)	Hematite, goethite		0.49	68.8	11.9	30.0	0.28	0.35	
Cherepovez district, Vologda, Russia									
PY (0-20)	Hematite, Al-goethite		0.77	62.7	5.4	21.8	0.20	0.11	0.40
B (31-52)	Hematite, Al-goethite		0.48	75.3	6.7	29.8	0.18	0.07	0.34
C (66-85)	Hematite, Al-goethite		1.18	63.0	11.3	28.8	0.28	0.35	0.63

Fe_{Tot} – total content Fe in soil, Fe_{DCB} – Fe, extracted with DCB, $I_{Lab-corr}$ – I_{Lab} index after correction for Al in goethite.

of hematite and goethite. Replacement of Fe^{3+} to Al^{3+} is taken into account through amendments to the color of goethite, admitting that Al-hematite color does not differ from color pure hematite.

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Validating new methods in luvisols is showed an agreement index values I_{Lab} with real [Hematite/(Hematite+Goethite)] in those parts where there are profile have pure hematite and goethite. In luvisols containing Al-goethite and Al-hematite, admixture Al generates an error in the calculation. To delete the error, it is proposed an amendment to the distorting effects of aluminum in lattice goethite, ewith high content: 8-12 mole Al %. Cambisols have a value index I_{Lab} also vary depending on the impurities Al in hematite and goethite. From samples with particles of pure goethite Red higher than cambisols containing Al-goethite. After adjustment for the Al-share in goethite the share of hematite becomes comparable to its share in the cambisols samples with pure goethite.

Acknowledgment

The author thanks N.P. Kirillova for help in the work.

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