

*Geophysics*

## **Intra-Annual and Seasonal Variations of Sub-Micron Aerosols Concentration and their Connection with Radon Content in Surface Boundary Layer of Tbilisi City**

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(Presented by Academy Member Tamaz Chelidze)

**ABSTRACT.** Results of the analysis of variations in sub-micron aerosols concentration with the diameter of  $\geq 0.1$  micron (LgN) and their connections with the radon content (Rn) in surface boundary layer of Tbilisi city (Georgia) are given. The data of daily mean values of the investigated parameters from 12.2009 to 11.2010 without taking into account the weather conditions (365 days, from 9 to 17-18 h) are analyzed. The special features of variations of radon and sub-micron aerosols according to annual and also winter, spring, summer and autumn data are studied. The effect of radon on the formation of sub-micron aerosols for indicated seasons are revealed. The changeability of sub-micron aerosols concentration and radon content at different seasons has the complex nature (intra-annual variations - tenth order polynomial for Rn and LgN, winter - linear regression for Rn and LgN, spring - fifth order polynomial for Rn and LgN, summer - linear regression for Rn and fifth order polynomial for LgN, autumn - sixth order polynomial for Rn and tenth order polynomial for LgN). The data of autocorrelation functions for the indicated time-series of observations are cited. The correlation and regression analysis of the connections between real values and residual components of time-series of LgN and Rn for the indicated seasons of year is carried out. Thus, direct correlation between radon content and sub-micron aerosol concentration for all seasons of year is observed. For real data the closest correlation is found in autumn, least close - in summer, for residual components – in winter and summer accordingly. © 2016 *Bull. Georg. Natl. Acad. Sci.*

**Key words:** radon, sub-micron aerosols

Atmospheric aerosol is the mixture of ordinary particles of the natural and anthropogenic origin (mineral aerosol, sea aerosol, the solid ejections of industrial enterprises and transport, etc.) and the so-called secondary aerosol. Secondary aerosol is formed in

the presence of the chemical and photochemical reactions according to the scheme of gas→particle. However, it turned out that radioactive and cosmic radiation contributes to the acceleration of the processes of the secondary aerosol formation.

In the work [1] it is shown that in Trombay (India, northeastern industrial suburb of Mumbai, 18.97° N, 72.83° E, 7 m above sea level), surface radon actively strengthens the processes of the formation of Aitken's nuclei in the atmosphere. This work did not turn itself a considerable attention. Later in the works [2, 3] the additional information about the influence of ionizing radiation on the fine dispersed aerosols formation was obtained. Our early studies [4-6] on the basis of an insignificant quantity of experimental data confirmed those results obtained in [1]. From 2009 the detailed studies of the effects of radon, gamma-radiation and cosmic radiation on the formation of sub-micron aerosols as one of the most important characteristics of smog in Tbilisi (Georgia) are carried out [7-9].

The special features of the effect of the radio nuclide emission in the formation of secondary aerosols in conditions of Tbilisi city (Tbilisi type of smog) are revealed. Intensification of the aerosol pollution of the atmosphere by the ionization (in essence of radon) under the conditions of Tbilisi is so strong that it leads to worsening of the air quality with respect to its ionic composition. In Tbilisi, smog is characterized by the impossible feedback of the radon content under the natural conditions, gamma and cosmic radiations with the concentration of light ions in air, caused by the formation of secondary aerosols, which in conjunction with the ordinary particles is capable of joining more ions to itself. Tbilisi type smog can occur also in other strongly contaminated cities and environments [8, 10-13].

This work is the continuation of the foregoing studies. The result of analysis of changeability of radon and sub-micron aerosols content at surface boundary layer of Tbilisi city, and also the effects of radon on the aerosols formation at different seasons are presented below.

## Materials and Methods

The radon content (Rn) was determined by the sampling method of air through the filter with the subse-

quent calculation of alpha particles of short-lived products at its decay. Content of a total quantity of sub-micron aerosols by diameter  $\geq 0.1 \mu\text{m}$  ( $\text{N}, \text{cm}^{-3}$ ) was measured by instrument FAN, which works in the counting regime. Radon and aerosol concentrations measurements were conducted 4 times a day at the height of the 3rd floor of the building of the cloud chamber of the Institute of Geophysics (8 meters above the level of soil, 41.754° N, 44.927° E, the height - 450 m above sea level), into 9, 12, 15 and 18 hour (in the winter time - 17 hours) [7,8,10-13]. The concentration of radon in correspondence with the method was determined [14].

The work gives the results of measurements from December 2009 to November 2010. The analysis of data is carried out without taking into account weather conditions. The data of the daily average values of the investigated parameters are analyzed (356 days, from 9 to 17-18 h).

In the proposed work the analysis of data is carried out with the use of the standard statistical analysis methods of random events and methods of mathematical statistics for the non-accidental time-series of observations [15].

The following designations will be used below: Rn – daily mean radon content in air ( $\text{Bq}/\text{m}^3$ ), LgN – logarithm of daily mean sub-micron aerosols concentration  $\text{N}$  ( $\text{cm}^{-3}$ ), Min – minimal values, Max - maximal values, Stdev - standard deviation,  $C_v$  - coefficient of variation (%),  $R_{\text{real}}$  and  $R_{\text{res}}$  - coefficient of linear correlation between real and residual time-series data of Rn and LgN accordingly,  $\alpha$  – the two-sided level of significance,  $R_a$  - coefficient of autocorrelation (lag=1 day) with  $\alpha \leq 0.05$ ,  $R^2$  – coefficient of determination,  $K_{\text{dw}}$  – Durbin-Watson statistic. The trend curve was determined by the optimum selection of the regression equation of the dependence of real data on time-series and Durbin-Watson statistic value for the residuals components (optimum combination values of  $R^2$  and  $K_{\text{dw}}$ ).

Five continuous time-series of average diurnal values of Rn and LgN were analyzed. Year – from

2009 December 1 to 2010 November 30 (in Fig. 1 the number of days are from 1 to 365); winter – from 2009 December 1 to 2010 February 28 (in Fig. 2 the number of days are from 1 to 90 days). For spring, summer and autumn are analogous.

The dimensionality of the investigated parameters is omitted further for convenience. In order not to overload the text the value of the coefficient Rn and LgN in the regression equations with days (polynomials of different degree) are also omitted.

## Results

In Table 1 statistical characteristic of daily mean values of Rn and LgN at investigation period is presented. As follows from Table 1 the values of indicated parameters in five periods change in the following ranges.

Rn: year and winter - from 0.7 to 13.1 (average values – 3.8 for year and 5.1 for winter); remaining seasons of year – from 0.7 (spring) to 12.3 (autumn) with average values from 2.5 to 4.7, accordingly. The greatest variation in the concentration of radon according to annual data is observed (52.7%), smallest - in summer (22.8%). In winter and autumn the values of  $C_v$  are approximately identical (45.8-45.5%). The greatest autocorrelation in time-series of annual data is observed (from 1 to 14 lags, with 16, from 21 to 23

and with 27 lags), smallest - for spring (with 1 lag only).

LgN: from 2.48 (year and winter) to 4.20 (year and summer) with average values from 3.15 (spring) to 3.44 (summer). The greatest variation of aerosol concentration is observed according to winter data (9.3%), smallest - in summer (8.1%). The greatest autocorrelation in time-series of annual data is observed (from 1 to 8 lags, with 18 and 21 lags), smallest - for spring (with 1 lag), in winter autocorrelation in time-series of LgN observations is absent. As a whole the time-series of observations of the aerosols is considerably less autocorrelated than series of radon observations.

The significant linear correlation between real data of Rn and LgN for all investigated periods of year is observed. The greatest correlation is observed in autumn (0.64), smallest - in summer (0.26).

In Table 2 information of parameters of changeability of radon and sub-micron aerosols content in air at different periods of year are presented. Figs. 1-5 depict the data of variations of real and calculated values of the concentration of radon and sub-micron aerosols in Tbilisi for five indicated periods of year.

As follows from Table 2 and Figs. 1-5 the changeability of radon and sub-micron aerosols concentration at different seasons of year has the complex na-

**Table 1. The statistical characteristics of radon and sub-micron aerosols content in air**

Parameter	Year	Winter	Spring	Summer	Autumn
	Radon				
Min	0.7	0.7	0.7	1.3	0.9
Max	13.1	13.1	6.0	5.3	12.3
Average	3.8	5.1	2.5	3.0	4.7
Stdev	2.0	2.3	0.9	0.7	2.1
$C_v$ , %	52.7	45.8	37.1	22.8	45.5
$R_a$ , lag	1-14,16,21, 22,23,27	1,6,7	1	1-2	1-6
	Sub-micron aerosols (LgN)				
Min	2.48	2.48	2.55	2.66	2.66
Max	4.20	3.94	3.71	4.20	4.12
Average	3.30	3.32	3.15	3.44	3.29
Stdev	0.30	0.31	0.27	0.28	0.28
$C_v$ , %	9.1	9.3	8.6	8.1	8.4
$R_a$ , lag	1-8,18,21	No	1	1-5	1-4
$R_{Real}$	0.42	0.51	0.42	0.26	0.64
$\alpha(R_{Real})$	<0.001	<0.001	<0.001	0.01	<0.001

**Table 2. Parameters of changeability of radon and sub-micron aerosols in air at different seasons of year**

Parameter	Year	Winter	Spring	Summer	Autumn
	Radon (Polynomial equation of the regression)				
Order	Tenth	Linear	Fifth	Linear	Sixth
R <sup>2</sup>	0.49	0.12	0.24	0.09	0.59
α (R <sup>2</sup> )	<0.001	0.001	<0.001	0.015	<0.001
K <sub>dw</sub>	1.48	1.63	1.6	1.53	1.52
α (K <sub>dw</sub> )	0.01	0.025	0.025	0.01	0.01
Sub-micron aerosols (Polynomial equation of the regression)					
Order	Tenth	Linear	Fifth	Fifth	Tenth
R <sup>2</sup>	0.25	0.03	0.12	0.37	0.19
α (R <sup>2</sup> )	<0.001	0.1	0.001	<0.001	<0.001
K <sub>dw</sub>	1.38	1.82	1.61	1.2	1.57
α (K <sub>dw</sub> )	0.01	0.05	0.025	<0.01	0.025
R <sub>res</sub>	0.48	0.63	0.43	0.30	0.39
α (R <sub>res</sub> )	<0.001	<0.001	<0.001	0.0015	<0.001

ture.

According to the data of year intra-annual variations of Rn and LgN has the form of the polynomial of the tenth power. The decrease from winter to spring and increase from summer to autumn is characteristic for radon concentration. Variations of the values of LgN during the year have more or less uniform nature, without the abrupt changes from one season to the next. Value of linear correlation coefficient between residual components of Rn and LgN is 0.48 (Table 1, Fig. 1).

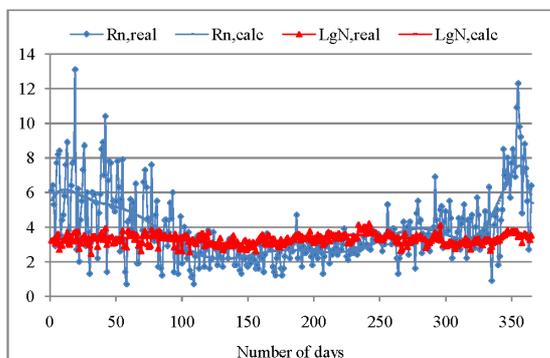
In winter variations of Rn and LgN are described by the linear regression equation, the considerably decreased values of Rn and a very weakly increased ones of LgN from 2009 December 1 to 2010 February 28. Value of R<sub>res</sub> between Rn and LgN is 0.63 (Table 1, Fig. 2).

In spring changeability of radon and sub-micron

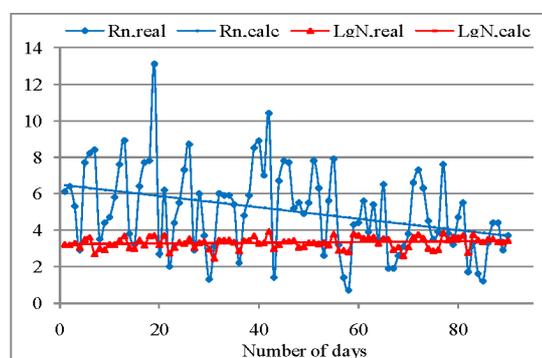
aerosol concentration has the form of the fifth order polynomial. Concentration of radon in the first half of March sharply decreases. Then undulating changes in the values of Rn are observed to the end of the spring. Values of LgN during the spring change undulating. Value of linear correlation coefficient between residual components of Rn and LgN is 0.43 (Table 1, Fig. 3).

In summer, changeability of Rn is described by the equation of linear regression and variations of LgN by the fifth order polynomial. A weak linear growth of radon concentration is observed for summer. During the undulations of the sub-micron aerosol concentration reaches its greatest values in the second half of July. Value of R<sub>res</sub> between Rn and LgN is 0.30 (Table 1, Fig. 4).

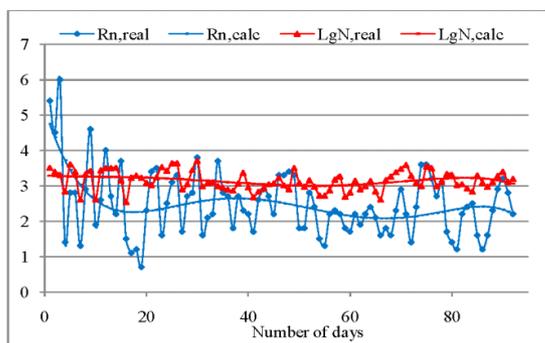
In autumn the variation of radon content in air by



**Fig. 1.** Intra-annual variations of radon and submicron aerosols content in air (real and calculate data)



**Fig. 2.** Variations of radon and sub-micron aerosols content in air in winter (real and calculate data)

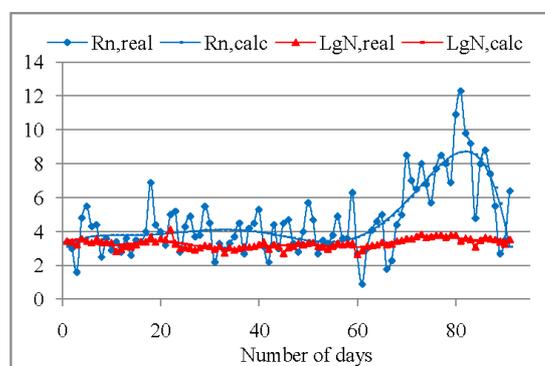


**Fig. 3.** Variations of radon and sub-micron aerosols content in air in spring (real and calculate data)

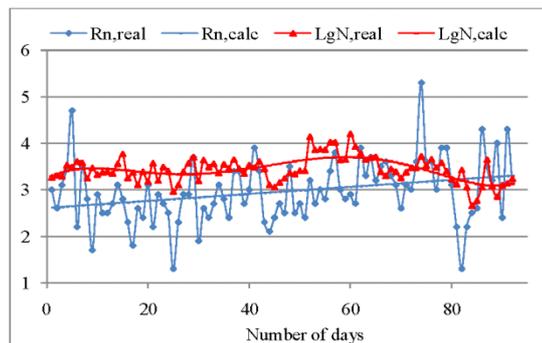
the sixth order polynomial and changeability of sub-micron aerosols concentration by the tenth order polynomial are described. During the undulations in September and October a sharp increase of radon concentration in the first half of November is noted. Then, toward the end of November - sharp decrease of Rn values. Sharp jumps in the fluctuation of LgN values were not observed. Value of linear correlation coefficient between residual components of Rn and LgN is 0.39 (Table 1, Fig. 5).

## Discussion

In the strongly polluted cities and localities the ionizing radiation (radon, cosmic rays and gamma radiation) contributes to the formation of secondary aerosols and with respect to an increase of air pollution. Main role in variations of the sub-micron aerosol values at surface boundary layer belongs to radon. This effect was discovered in Tbilisi for all periods of



**Fig. 5.** Variations of radon and sub-micron aerosols content in air in autumn (real and calculate data)



**Fig. 4.** Variations of radon and sub-micron aerosols content in air in summer (real and calculate data)

year with all weather conditions, including days with the photochemical smog, fogs, etc. Weaker correlation between Rn and LgN is observed in summer.

The time-series of observations of the aerosols is considerably less auto correlated than series of radon observations.

In some periods of the year (year, winter and spring) the time variation of the radon and sub-micron aerosol concentration has similar nature and is described by the identical equations of regression. In summer and in autumn the nature of the time variations of the radon and sub-micron aerosols concentration is different.

Correlation between residual components of Rn and LgN as for their real values for all periods of year with all weather conditions observed also. The latter confirms the presence of the close connection between the content of radon and sub-micron aerosols.

However, there appears the problem of the separate determination of the ranges of sizes and concentration of the secondary aerosols, formed under the action of the different kind ionizing radiation with different state of atmosphere (temperature, humidity, solar radiation, the forming secondary aerosols gases, etc.).

Further detailed field and laboratory investigations of this effect with the use of equipment for measuring the concentration of aerosols over a wide range of the spectrum of their sizes are also desirable.

Besides atmospheric industrial and environmental electricity and ecological aspects of the indicated effect of ionizing radiation it is important also to esti-

mate its direct and indirect climatic effects (influence on the solar radiation, visibility, cloudiness, precipitation, thunderstorm activity, etc.).

### Conclusions

Some special features of time variations of radon and sub-micron aerosols concentration in air under the conditions of Tbilisi city for different periods of year are revealed.

The strong effect of catalyzation of the processes of secondary aerosol formation at surface boundary layer of atmosphere according to the scheme of gas'! particle by the ionizing radiation is observed.

Subsequently the continuation of the indicated works is provided. In particular, the study of the direct and indirect influence of radon and other ionizing substances on climate changes causes interest.

### გეოფიზიკა

## სუბმიკრონული აეროზოლების კონცენტრაციის ყოველწლიური და სეზონური ვარიაციები და მათი კავშირი რადონის შემცველობასთან ქალაქ თბილისის მიწისპირა სასაზღვრო ფენაში

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ნაშრომში განხილულია სუბმიკრონული აეროზოლების დიამეტრით  $\geq 0,1$  მიკრონი (LgN) კონცენტრაციის ვარიაციის ანალიზის შედეგები და მათი კავშირი რადონის შემცველობასთან (Rn) ქ. თბილისის მიწისპირა სასაზღვრო ფენაში. გაანალიზებულია განსახილველი პარამეტრების დღიური საშუალო მნიშვნელობები 12.2009-დან 11.2010-მდე ამინდის პირობების გათვალისწინებლად (365 დღე, 9 სთ-დან 17-18 სთ-მდე). შესწავლილია რადონისა და სუბმიკრონული აეროზოლების ვარიაციის განსაკუთრებული თავისებურებები წლიური მონაცემების მიხედვით, ასევე ზამთრის, გაზაფხულის, ზაფხულისა და შემოდგომის პირობებში. გამოვლენილია რადონის ზეგავლენა სუბმიკრონული აეროზოლების წარმოქმნაზე სეზონის მიხედვით. რადონის შემცველობისა და აეროზოლების კონცენტრაციის ვარიაციას სხვადასხვა სეზონზე აქვს კომპლექსური ხასიათი (შიდა-წლიური ვარიაციები - Rn-ისა და LgN მათე რიგის პოლინომი, ზამთარში - Rn-ისა და LgN-ის წრფივი რეგრესია, გაზაფხულზე - Rn-ისა და LgN მჭიდრო რიგის პოლინომი, ზაფხულში - Rn-ისა და LgN-ის წრფივი რეგრესია, შემოდგომაზე - Rn-ის მკვეთრი რიგის პოლინომი და LgN-ის მათე რიგის პოლინომი). მოცემულია დაკვირვების მონაცემების აუტოკორელაციის ფუნქციის მნიშვნელობები აღნიშნული პერიოდისთვის. ჩატარებულია Rn-ისა და LgN-ის დროითი მწკრივების

რეალურ მონაცემებსა და ნარჩენ კომპონენტებს შორის კავშირის კორელაციური და რეგრესიული ანალიზი აღნიშნული სეზონებისათვის. ამრიგად, გამოვლენილია პირდაპირი კორელაცია რადონის შემცველობასა და სუბმიკრონული აეროზოლების კონცენტრაციას შორის წელიწადის ყველა სეზონისათვის. ნაპოვია რეალური მონაცემებისათვის ყველაზე ახლო კორელაცია შემოდგომაზე, ნაკლებად ახლოზაფხულში, ნარჩენი კომპონენტებისათვის - ზამთარსა და ზაფხულში.

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