Estimate for Radiative Forcing of Smoke Aerosol from 2010 Summer Fires Based on Measurements in the Moscow Region

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Received March 28, 2013; in final form, September 25, 2013

Abstract—Temperature and radiation effects of the 2010 summer fires are estimated on the basis of measurements at the Zvenigorod Scientific Station (ZSS) of the Obukhov Institute of Atmospheric Physics, Russian Academy of Sciences (RAS), in the Moscow region. The surface air temperature during the 2010 summer smoke varied in antiphase with the aerosol mass concentration, and the thermal radiation balance in the surface layer of the atmosphere varied in phase. Under extreme smoke of the surface layer in August 2010, the reduction in surface air temperature at ZSS has been found to reach 4 K with an increase in the downward flux of thermal radiation by an average of 20 W/m² and a decrease in the difference between upward and downward fluxes of thermal radiation by an average of 24 W/m².

Keywords: aerosol forcing, wildfires, surface aerosol, trajectory analysis

DOI: 10.1134/S0001433814020091

INTRODUCTION

Large wildfires lead to smoke in the air basin over vast areas and changes in the radiation balance of entire regions. Extreme smoke in the atmosphere over the European territory of Russia was registered during the 2010 summer fires associated with the prolonged blocking of zonal transport in the mid-latitude troposphere [1]. Smoke aerosol reduces the influx of solar radiation; as a result, the air temperature in the surface layer in smoky days decreases compared to the temperature in no-smoke conditions. The cooling of the atmospheric surface layer due to extreme 2010 summer smoke was estimated in [2] using atmospheric aerosol parameters measured at the Zvenigorod Scientific Station (ZSS) of the Obukhov Institute of Atmospheric Physics, Russian Academy of Sciences (IAP RAS) (55.7° N, 36.8° E). Here, model calculations were used for aerosol radiative forcing.

Direct measurements of aerosol forcing and temperature variations caused by excess aerosol content in the atmosphere are complicated by the necessity of a number of conditions to be satisfied simultaneously. First, against the background of general smoke, a time interval (base period) is needed with an almost background content of atmospheric aerosol, which can be used as a reference for estimating the surface air cooling due to increased aerosol content. Second, the modes of the base period (day) and smoke days must refer to the same air mass because the surface air temperature can significantly vary with changed air masses even if no aerosol effects are available. Third, both the base day and the days with increased aerosol concentrations should be fair with some clouds.

To what extent did the extremely strong smoke in the surface air of the Moscow region in August 2–9, 2010, satisfy the abovementioned necessary conditions? During this period of inactive anticyclone without air mass change, the polluted air is transported from areas of forest and peat fires, the largest sources of which were located east and southeast of ZSS. On August 5, 2010, the surface air at ZSS was purified (without air mass change) and the aerosol content decreased to a level close to the characteristic local level of this time of year. It should be noted that, under anticyclonic conditions, the extreme smoke of August 2–9, 2010, involved almost no cloudiness (except for August 3, 2010).

Thus, in the summer of 2010, the Moscow region was characterized by conditions allowing one to directly estimate the effects of atmospheric smoke associated with fires. This paper estimates the temperature and radiation effects of the 2010 summer fires on the basis of measurements conducted at ZSS.

INSTRUMENTATION AND MEASUREMENT TECHNIQUE

All measurements were conducted in the near-surface layer of the atmosphere (2–15 m). From June 21, 2010, to August 26, 2010, detailed measurements of the downward flux of thermal radiation in the wavelength range 3.5–50 µm, air temperature, and aerosol number and mass concentrations in the size range...
0.3–10.0 µm were conducted at ZSS. From August 2 to 10, 2010, samples of aerosol particles with a diameter in the range from 0.7 to 1.0 µm were collected and their chemical composition was analyzed by infrared spectroscopy.

The measurements of aerosol number concentration were performed at a height of 2 m above the surface a few times during daylight hours using an AZ-10-0.3 standard calibrated photoelectric aerosol particle counter. The aerosol number concentrations were measured in six particle size (diameter) ranges: 0.3–0.4, 0.4–0.5, 0.5–1.0, 1.0–2.0, 2.0–5.0, and 5.0–10.0 µm, which makes it possible also to analyze the aerosol particle size distributions. The relative error of measured number concentrations was 20% for all particle size ranges.

The AZ-10-0.3 particle counter allows one to quickly and efficiently convert number concentrations into the standard mass indicators of aerosol content in the air PM1.0 (particles with sizes <1.0 µm), PM2.5 (<2.5 µm), PM5.0 (<5.0 µm), and PM10 (<10.0 µm). During the conversion of the number concentration into the mass concentration, the specific density of smoke aerosol was taken to be 1.05 g/cm³ (close to the density of tar oil, pitch, and rosin).

To measure the aerosol concentration, a sample of air of a volume of 5 dm³ was collected, which is enough to obtain under smoke conditions a statistically stable number concentration even for the largest fraction (5–10 µm) of particles (the least in the air). Under conditions of extreme smoke, when the total number concentration of smoke aerosol exceeds the upper limit of the measured particle counter (5 × 10³ dm⁻³), the samples were diluted (in the ratio of 1 : 5) by clean air obtained by AFA-VP filters.

Continuous measurements of the downward flux of thermal radiation DLR (W/m²) in the wavelength range 3.5–50 µm were conducted at a height of 15 m from the surface with 1-min averaging over the whole period of analysis using an Eppley PIR automated infrared radiometer. This infrared radiometer was used also to record the thermal radiation balance B = DLR – ULR, where DLR and ULR are the downward and upward fluxes of thermal radiation at the instrument level, respectively. The error of DLR and B measurements was 3 W/m².

The measurements of surface air temperature \( T (°C) \) were conducted with 1-min averaging over the whole period of analysis by temperature sensors located close to the infrared radiometer also at a height of 15 m above the surface. The error of \( T \) measurements was 0.1 K.

The absolute humidity of air \( Ah (g/m³) \) was measured on the basis of temperature and relative humidity measured at a height of 2 m above the surface with the help of standard meteorological instruments at the ZSS meteorological station.

Using a cascade impactor, aerosol particles of sizes 0.7–1.0 µm were deposited on a germanium substrate, and the chemical composition of a deposited aerosol was determined by IR spectroscopy. The air intake was carried out at a height of 3 m above the surface.

The analysis was based on averaging over 12 daylight hours (from 07:00 to 19:00) of surface air temperature \( T \), downward flux of thermal radiation DLR, thermal radiation balance B, aerosol concentration PM10, and absolute air humidity Ah.

The HYSPLIT model [3, 4] was used to calculate the back trajectories of air masses for each daylight hour (from 07:00 to 19:00 local time) of each day of August 2–10, 2010, at different heights (with a vertical step of 100 m) in the atmospheric boundary layer up to 1 km (a total of 1188 trajectories). All back trajectories for this height range were analyzed jointly within a single day: one should take into account the air inflow not only into the near-surface layer, but also for the whole layer of active mixing, the height of which in the smoke days at ZSS on the average was around 1 km [2]. On the basis of arrays of back trajectories of air masses for each day of August 2–10, 2010, we constructed the spatial distributions (maps) of the probability of back trajectory passing with a resolution of 0.25° in latitude and longitude.

**RESULTS**

The smoke in the surface air, which was detected at ZSS from July 20 to August 19, 2010, involved three stages separated by two changes of air masses with a surface layer cleaning. The first (from July 20 to August 1) and third (from August 10 to 18) stages were characterized by increased levels of smoke (I and III in Fig. 1). Extreme smoke at ZSS was recorded on August 2–9 (stage II in Fig. 1) for the same air mass. The subsequent analysis refers specifically to the second stage smoke. Maximum values of PM10 (around 0.6 mg/m³) were recorded on August 8, with a subsequent decline of smoke. At the same time, there was a single day (August 5) during the second stage of smoke when PM10 dropped almost to typical local values (0.01–0.02 mg/m³) for this season (see Fig. 1). The aerosol optical thickness (AOT) averaged over daylight hours at a wavelength of 550 nm (\( \tau \)) on this day did not exceed 0.5, reaching 4 or more in the days of smoke [2, 5]. In this case, the background values of \( \tau \) are normally no more than 0.2 for the summer season.

According to the results of the trajectory analysis, the air masses arriving during daylight hours of August 5, 2010, into the atmospheric layer above ZSS contained a small amount of aerosols because previously they rounded from the southwest the zone of nearest large forest and peat fires located east and southeast of ZSS. It can be seen from Fig. 2 that the trajectories of this day, like in the remaining days from August 2 to 10, are twisted clockwise, which is typical for an anticyclonic circulation. Figure 2 shows the spatial distributions of
the probability of passing of back trajectories of air masses that came to the ZSS area within the 12 daylight hours each day from August 2 to 10, 2010. A comparison between Figs. 1 and 2 shows that, in the days of extreme smoke (August 7–9, 2010), the higher the probability of back trajectory passing in the area of nearest fires in the south east of Moscow is, the higher the aerosol mass concentration at ZSS is.

According to the measurements at ZSS on August 2 to 9, 2010, the distributions of aerosol number concentration by particle size had a maximum between 0.4 and 0.5 µm, while the corresponding typical distributions had a maximum near 0.3 µm (see Fig. 3a). The distribution maximum between 0.4 and 0.5 µm is typical for the pyrolysis of wood and peat [6, 7]. The distribution functions of the mass concentration by particle sizes during the smoke days had maximums between 0.4 and 1.0 µm (see Fig. 3b). The increased content of large submicron particles can explain the reddish color of the smoky sky in the days of extremely high aerosol concentrations.

During the pyrolysis of wood and peat, the deposited aerosol is a black or brown resinous substance [7]. The samples collected on August 2–9, 2010, also contained a brown—black resinous substance. In this case, the samples related to the peak of smoke on August 6 to 9 were a single drop of a resinous substance [8]. Aerosol particles in the size range of 0.7–1.0 µm could be microdrops of a tar-type resinous substance with an admixture of black carbon (BC) and inorganic components. Since coarse submicron particles are the largest contributors to PM10, the specific density of smoke aerosol during the conversion of number concentration into mass concentration was taken equal to the average specific density for tar oil, pitch, and rosin.

According to IR spectrometry data, the samples of deposited aerosol particles had an increased content of BC and carboxylic acids (functional group—COOH), which is also typical for the pyrolysis of wood [9]. The maximum content of carboxylic acids in these samples falls on the days of extreme smoke from August 6 to 9, 2010.

Since the functional group—COOH is hydrophilic, the aerosol particles containing carboxylic acids can become hygroscopic. In this case, during a growth of humidity (for example, at night) or air cooling during its uplift, the smoke particles could be activated, growing in size. For BC particles in the presence of hydrophilic organic substances, one can expect that their scattering and absorption cross sections should increase [10]. Therefore, activated smoke particles could absorb short-wave radiation more efficiently than purely BC particles. The fog formed at night on hygroscopic smoke particles could prevent radiative cooling of the surface air layer, as well as contribute to air warming due to the release of the latent heat of water vapor condensation.

Under anticyclonic conditions during the long-term blocking on August 2 and from August 4 to 9, 2010, almost no cloudiness was available, and the daily mean temperature of the surface air in the “clear” day of August 5, 2010, can be considered a base day against which one can estimate the cooling in more “hazy” days. A similar comparison can be made for daily mean values of the downward flux of thermal radiation.
and thermal radiation balance at a level of 15 m above the surface.

Table 1 shows the daily mean anomalies of $T$, DLR, and $B$ in cloudless smoke days of August 2–9, 2010, with respect to the “clean” day of August 5, 2010.

To determine the true cooling of surface air in smoke days with respect to no-aerosol conditions ($\tau = 0$), it is necessary to take into account the air cooling on August 5 at $\tau = 0.5$. This can be estimated by a linear regression bias for the dependence of $\Delta T$ on $\tau$.

The linear regression for the dependence of $\Delta T$ on $\tau$ (the correlation coefficient is $r = -0.89$) from the data presented in Table 1 has the form $\Delta T(\tau) = 0.7 - \tau$ for the estimation error in the bias of linear regression $\delta_T = 0.4$ K. The corresponding cooling of the surface air on August 5 at ZSS with respect to conditions at $\tau = 0$ and with measurement errors and estimates of linear regression bias was $-0.7 \pm 0.5$ K. Similarly, the linear regression for the dependence of DLR on $\tau$ ($r = 0.31$) has the form $\Delta \text{DLR}(\tau) = 7 + 3\tau$ ($\delta_{\text{DLR}} = 8 \text{ W/m}^2$) and, for the dependence of $\Delta B$ on $\tau$ ($r = 0.87$), $\Delta B(\tau) = 9\tau$ ($\delta_{\text{AB}} = 5 \text{ W/m}^2$). For $\tau = 0$, the biases of
\( \Delta N/\Delta d, \text{dm}^{-3} \mu m^{-1} \) (a)

- 02.08.2010
- 03.08.2010
- 04.08.2010
- 06.08.2010
- 07.08.2010
- 08.08.2010
- 09.08.2010
- 05.08.2010

\( \Delta M/\Delta d, \mu g \text{dm}^{-3} \mu m^{-1} \) (b)

- 02.08.2010
- 03.08.2010
- 04.08.2010
- 06.08.2010
- 07.08.2010
- 08.08.2010
- 09.08.2010
- 05.08.2010

Fig. 3. Distribution of aerosol number concentration (a) and mass concentration (b) by particle sizes in August 2010 at ZSS. The values of the distribution functions correspond to the left limits of particle size ranges marked by dashed lines.

\( \Delta \text{DLR} \) and \( \Delta \text{B} \) with respect to background conditions for ZSS (in accordance with the appropriate measurement errors and estimates for linear regression bias) were 7 (±11) W/m² and 0 (±8) W/m², respectively.

Table 2 shows the corrected anomalies of \( T \), \( \text{DLR} \), and \( \text{B} \) in the smoke days of August 2–9, 2010, with respect to completely no-aerosol conditions and with appropriate measurement errors and linear regression bias estimates.

According to Table 2, the maximum anomaly in surface air temperature at ZSS during smoke could reach values of around −4 K.

By estimates of [2] and on the basis of measurements and model calculations of shortwave radiative
forcing, the anomaly of 12-h average surface air temperature reached –6 K due to aerosol under conditions of extreme atmospheric smoke on August 7–9, 2010. The difference in the estimates of maximum temperature anomalies due to aerosol cooling in [2] from those obtained in this study is explained by the fact that the estimates in [2] are obtained by a decreased flux of incoming solar radiation without a proper analysis for the flux of thermal radiation. Becoming hygroscopic in the presence of hydrophilic carboxylic acids, the smoke aerosol not only reduced the solar radiation, but also could efficiently absorb shortwave and near-infrared radiation due to activated particles, and it could form (under wetting conditions) hazes that partially inhibit the outgoing longwave radiation.

From August 2 to 9, 2010, the daily average surface air temperature \( T \) and the PM10 aerosol concentration varied almost in the antiphase (see Fig. 4a) with a correlation coefficient \( r = -0.67 \). The reduction of PM10 on August 2, 5, and 9 led to an increase in surface air temperature in these days, while the increase of PM10 on August 4 and August 6–8 caused a decrease in surface air temperature in the respective days. In this case, the variation of thermal radiation balance and PM10 concentration varied in phase \( r = 0.77 \), see Fig. 4a): the higher the PM10 aerosol mass concentration is, the smaller the difference between downward and upward fluxes of thermal radiation is. This speaks about the absorption of downward radiation by smoke aerosol in the visible and near-infrared ranges [6, 7] and the possible greenhouse effect of smoke aerosol.

The downward flux of thermal radiation DLR decreased with increasing PM10 on August 6–9, 2010, but was significantly higher than in the relatively clean atmosphere of August 5 (see Fig. 4b), although the surface air on August 5 was warmed stronger. This can be linked also with a possible greenhouse effect by smoke aerosol. With increasing PM10, the absolute humidity \( \Delta h \) increased slightly (1.4 g/m\(^3\) or by 10%), which is possibly caused by the formation of water vapor under pyrolysis and the combustion of wood and peat. The water-vapor content in the bulk changed likewise slightly (from 2.0 to 2.5 cm, or by 25% [5]). The water-vapor content in the bulk (according to [5]) and \( T \) were weakly correlated, which may indicate that the contribution of the influence of thermal radiation absorption by water vapor in the bulk on the change in surface air temperature in smoke days is negligible.

It should be noted that the moisture content of the atmosphere under anticyclonic conditions during the 2010 summer fires was significantly higher than during the 1972 and 2002 summer fires in the European part of Russia [11–14].

### Table 1. Changes in surface air temperature (\( \Delta T \)), downward flux of thermal radiation (\( \Delta \text{DLR} \)), and thermal radiation balance (\( \Delta B \)) in the days of strong smoke with respect to the base day of August 5, 2010

<table>
<thead>
<tr>
<th>Date</th>
<th>( \tau )</th>
<th>( \Delta T, K )</th>
<th>( \Delta \text{DLR}, \text{W/m}^2 )</th>
<th>( \Delta B, \text{W/m}^2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>02.08.2010</td>
<td>1.0</td>
<td>–0.5</td>
<td>17</td>
<td>–16</td>
</tr>
<tr>
<td>04.08.2010</td>
<td>1.7</td>
<td>–0.9</td>
<td>7</td>
<td>–11</td>
</tr>
<tr>
<td>05.08.2010</td>
<td>0.5</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>06.08.2010</td>
<td>1.7</td>
<td>–0.1</td>
<td>25</td>
<td>–21</td>
</tr>
<tr>
<td>07.08.2010</td>
<td>2.6</td>
<td>–1.5</td>
<td>18</td>
<td>–25</td>
</tr>
<tr>
<td>08.08.2010</td>
<td>3.0</td>
<td>–2.5</td>
<td>12</td>
<td>–27</td>
</tr>
<tr>
<td>09.08.2010</td>
<td>2.4</td>
<td>–1.9</td>
<td>9</td>
<td>–20</td>
</tr>
</tbody>
</table>

The values of AOT \( \tau \) are based on data from [2].

### Table 2. Corrected anomalies of surface air temperature (\( \Delta T \)), downward flux of thermal radiation (\( \Delta \text{DLR} \)), and thermal radiation balance (\( \Delta B \)) due to the presence of aerosol in the atmosphere

<table>
<thead>
<tr>
<th>Date</th>
<th>( \Delta T, K )</th>
<th>( \Delta \text{DLR}, \text{W/m}^2 )</th>
<th>( \Delta B, \text{W/m}^2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>02.08.2010</td>
<td>–1.2 (±0.5)</td>
<td>24 (±11)</td>
<td>–15 (±8)</td>
</tr>
<tr>
<td>04.08.2010</td>
<td>–1.6 (±0.5)</td>
<td>14 (±11)</td>
<td>–11 (±8)</td>
</tr>
<tr>
<td>05.08.2010</td>
<td>–0.7 (±0.5)</td>
<td>7 (±11)</td>
<td>0 (±8)</td>
</tr>
<tr>
<td>06.08.2010</td>
<td>–0.8 (±0.5)</td>
<td>32 (±11)</td>
<td>–21 (±8)</td>
</tr>
<tr>
<td>07.08.2010</td>
<td>–2.2 (±0.5)</td>
<td>25 (±11)</td>
<td>–25 (±8)</td>
</tr>
<tr>
<td>08.08.2010</td>
<td>–3.2 (±0.5)</td>
<td>19 (±11)</td>
<td>–26 (±8)</td>
</tr>
<tr>
<td>09.08.2010</td>
<td>–2.6 (±0.5)</td>
<td>16 (±11)</td>
<td>–19 (±8)</td>
</tr>
</tbody>
</table>

Figure 5a shows the variations of thermal radiation balance \( B' \) (with 120-min moving averaging) for each cloudless day from August 2 to 9, 2010. In both morning and evening hours, \( B' \) is higher on smoke days than for the relatively clean base day of August 5. The reduced difference between downward and upward fluxes of thermal radiation in morning and evening hours in smoke days can be associated with a more efficient radiative heating of upper layers of the smoky air in comparison with its lower layers. In morning and evening hours, when the sun is low and the optical thickness of the smoky atmosphere is large, the radiation flux at the surface is considerably reduced. The more efficient heating of the air layer above the IR-radiometer level during all daylight hours in smoke days in comparison with its heating on the base day is confirmed also by the higher values of the downward flux of thermal radiation on smoke days (see Fig. 5b).
Fig. 4. Variations of PM10 concentrations in August 2010 at ZSS in comparison with variations in surface air temperature $T$ and thermal radiation balance $B$ (a), as well as with variations in the downward flux of thermal radiation DLR and absolute air humidity Ah (b).

The more uniform (by height) radiative heating of air in daytime hours of smoky days can be associated with reduced convection in comparison with the base day. It should be noted that, under anticyclonic conditions of blocking, the convective processes are generally suppressed. Under conditions of less developed convection, the magnitude of temperature and wind-speed pulsations is smaller. Figure 6a shows linear approximations of power spectra of surface air temperature pulsations in the frequency range from 0.01 to $1 \text{ min}^{-1}$ for August 2, 2010, and for each day of August from 4 to 9, 2010. According to Fig. 6a, the...
surface air temperature pulsation rate in smoky days at all frequencies of the specified frequency range is lower than for the relatively clean day of August 5. The reduction of convective processes in smoky days contributed to reduced heat removal from the surface level of the air and a weakened effect of surface layer cooling.

Figure 6b shows smoothed (with 120-min moving averaging) variations in surface air temperature $T'$ for each day of August 2 to 9, 2010. On the morning of smoky days, the surface air temperature increased with a lag of around 1.5–2 h relative to the course of temperature on the relatively clean day of August 5. This
can be associated with a weaker surface warming at high optical thicknesses of the smoky atmosphere at low sun altitudes at morning hours. The expected advance of the evening temperature drop in smoky days (due to a similar decrease in surface heating at large optical thicknesses of the smoky atmosphere at low sun altitudes at evening hours) was not revealed. This may point to a more efficient delay of outgoing heat by the surface layer of air in smoky days.

Figure 7 shows variations in daily mean PM10 aerosol concentration and averaged (over 12 h of evening and night from 07:00 to 07:00) surface air temperature $T$ of the next night. According to Fig. 7, the surface air temperature in three (cloudless) nights following the days of maximum smoke from August 6 to 8, 2010, became stable at around 21°C, which is almost 1°C above the surface air temperature during the night after the base day of August 5. This can be associated with the fact that, being activated under high humidity conditions at night, the hygroscopic smoke aerosol contributed to the reduction of night cooling rate of the surface layer of air. In this case, the temperature data for cloudy nights after August 2, 4, and 9 were not considered in view of possible distortions in respective estimates.

CONCLUSIONS

During the extreme smoke of the surface air in August 2010, the aerosol mass concentration reached 0.6 mg/m³, with the largest contribution to the mass of aerosol particles with sizes from 0.4 to 1.0 μm.

According to IR-spectrometry data on deposited aerosol, particles with sizes of 0.7–1.0 μm contained hydrophilic carboxylic acids and black carbon.

The extreme smoke of the surface air on August 7–9, 2010, was characterized by a decrease in the surface air temperature at ZSS by an average of 3 K and up to 4 K at a maximum cooling, with an increase in the downward flux of thermal radiation by an average of 20 W/m² (with a maximum increase of 36 W/m²) and a decrease in the difference between the downward and upward fluxes of thermal radiation by an average of 24 W/m² (with a maximum reduction of 36 W/m²).

The aerosol mass concentration and surface air temperature in the days of extreme smoke in August 2010 varied in the antiphase (with a correlation coefficient $r = -0.67$). The thermal radiation balance in the surface air layer and the PM10 mass concentration varied coherently (with a correlation coefficient $r = 0.77$).

In the smoky days of August 2, 4, and 6–9, the spectral power pulsations of surface air temperature in
the frequency range from 0.01 to 1 min\(^{-1}\) was lower than in the relatively clean day of August 5, which may be caused by weakened convection due to the reduction in the difference of heating between surface and higher layers during daylight hours.

The nocturnal surface air temperature after days with a smoke peak from August 6 to 8, 2010, was higher than at the night after the relatively clean day of August 5, 2010. This may indicate that there is a decrease in the rate of radiative cooling of the surface air layer by activated (under high humidity conditions in nighttime) hygroscopic smoke aerosol.

ACKNOWLEDGMENTS

This study was supported by the Russian Academy of Sciences; the Russian Foundation for Basic Research, project no. 12-05-00938-a; and the President of the Russian Federation, grant NSh-5467.2012.5.

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Translated by V. Arutyunyan