

Complex Assessment of Atmospheric Air Quality in the City of Gelendzhik

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Abstract—The atmospheric air quality is determined by the concentrations of some gaseous pollutants and mass concentrations of aerosol particles of different sizes. A wide range of atmospheric pollutants in both gaseous and aerosol phases was studied in the vicinity of Gelendzhik in July 2009, simultaneously at several land sites, in the water area of the bay, and at altitudes of up to 2200 m. No such complex experiments were carried out in that region before. The following characteristics of the atmospheric aerosol (3 nm–32 μm in size) were studied: elemental composition of particles (23 chemical elements) and concentrations of polyaromatic hydrocarbons (14 compounds), unsaturated hydrocarbons, total protein, biotoxins, and culturable microorganisms. The concentration fields of different air pollutants and the complex air pollution index were constructed using mathematical models of pollutant propagation and data on the hydrometeorological conditions during the period of measurements. The sources of aerosols in the region were detected from the study of the chemical composition of airborne particles. The results allowed us to estimate air pollutants and to calculate the complex air pollution index for the Gelendzhik area. The daily average concentrations of all the pollutants were compared to the daily average maximum permissible concentrations. All these concentrations were less than daily average maximum permissible concentrations. The complex air pollution index did not exceed 1. Hence, the air in the vicinity of Gelendzhik did not contain any significant pollutants in the period under study.

Keywords: air pollution, air quality, aerosol chemical composition, aerosol biological composition, aerosols sources, PM_x

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INTRODUCTION

Gaseous and aerosol pollutants and their variability in the atmosphere are currently studied all over the world [1–6], in view of their influence on atmospheric processes and the climate [7–10] and possible negative effect on ecosystems and human health [11–15]. Environmental protection agencies permanently toughen requirements for atmospheric air quality with the aim of improving living conditions [16–19]. Such gases as CO, CO₂, O₃, NO_x, and SO₂ and aerosol particles no larger than x (PM_x) are controlled the most often in different countries [1, 2, 20–24]. Earlier, sanitary norms were set only for PM₁₀ (i.e., mass of these particles the excess of which provokes the development of different diseases) (e.g., [16]). Recent research [11, 25] has shown that small particles affect

human health the strongest; therefore, masses of PM_{2.5}, PM₁, and even smaller particles are measured [23, 25–27] and hygienic norms are set for these quantities [16, 21, 28, 29].

It is evident that there are many more substances that negatively affect human health than we mentioned above. Their effect can be estimated via risks of developing different diseases under high concentrations of the substances in ambient air. Maximum permissible concentrations (MPC) of substances in air, that is, the concentrations which do not cause negative consequences yet, are used for such estimation in Russia [29–32].

It is impossible today to measure concentrations of all gaseous and aerosol (or adsorbed on the aerosol particle surface) atmospheric pollutants [1, 33–36].

Such measurements should be carried out in real time, since the gas-aerosol system under study is time dependent. In addition, flows of substances, which could be still transiting from one phase to another (evaporation, condensation, and nucleation) and participating in different chemical reactions, enter/exit a fixed volume [34, 36, 37].

There are simultaneously many different pollutants in air. Therefore, it is important to estimate the complex effect of the pollutants on human health. This problem has not been solved yet. Data on the combined effect of at least a pair of pollutants are lacking in the literature [35, 38]; such data are practically lacking for a larger number of pollutants. It is unclear whether synergism or antagonism of the pollutant effects exists, and if yes, how their effects intensify or weaken. Therefore, we do not consider the mutual effect of pollutants in this work.

Following [29], the linear *complex air pollution index* (API) Y_n can be defined as a sum of the mean concentrations of n different substances normalized to their daily mean MPC:

$$Y_n = \sum_{i=1}^n Y_i = \sum_{i=1}^n \left(\frac{q_{mi}}{\text{MPC}_{\text{dmi}}} \right)^{c_i}, \quad (1)$$

where Y_i is the single pollution index for the i th substance and q_{mi} is its mean concentration; c_i is the dimensionless constant of reduction of the harmful index of the i th substance to the harmful index of sulfur dioxide, it depends on the hazard category of this substance [29].

API serves the quantitative measure of the atmospheric air quality. The smaller Y_n , the less dangerous was the atmospheric air during the day.

It is evident that MPC_{dm} is not the only normalizing factor. The Y_i index can be estimated for both shorter and longer time periods. However, Russian regulations [30, 31] usually deal with the daily mean MPC. MPC for other periods have been found for a few substances [30, 31].

MPC_{dm} can be a benchmark for estimation of the well-being of the atmospheric environment. However, the use of this parameter as a measurement unit involves significant difficulties [39]. First, this parameter has not yet been determined for many pollutants. Second, even if one knows the concentration of, e.g., a heavy metal, the chemical compound of this element is unknown, and MPC_{dm} differs for different compounds of a metal. Third, daily mean MPC “miss” short-term emissions of high concentrations of a pollutant. To compare the data on air pollution for several substances, Y_n should be calculated for the same number (n) of impurities. If the n are different, then Y_n is calculated over Y_i indices of those five substances for which n is maximal [39].

Despite the urgency of such studies, no comprehensive assessments of API for gaseous pollutants and detailed assessments of aerosol pollutants have been carried out in a Russian region. The assessment of the Y_n and Y_i indices during several summer days of 2009 for Gelendzhik is the aim of this work.

MATERIALS AND METHODS

Gelendzhik is a resort city on the Black Sea coast of Russia with a population of about 100 thousand people, where the number of vacationers reaches two million in summer. There is no industry in the city; therefore, the main sources of pollution here should be long-distance transport of gas and aerosol pollution and local aerosol sources: automobile and sea transport, cooking, soil, road dust, vegetation, and seawater.

The complex API was assessed in summer 2009 in Gelendzhik using three ground-based observation posts (points Laboratory (L), Pier (P), and Cape (M) in Fig. 1), a research vessel (C) in the Gelendzhik Bay water area, and an aircraft laboratory flying over the coast at altitudes from 500 to 2200 m. Data on the hydrometeorological situation during the experiments were recorded at two weather stations (L and a station of the Institute of Oceanology (I)) with the help of meteorological complexes Vantage Pro2 Plus (Davis, Italy). The values of air pressure, temperature, relative humidity, solar radiation, wind speed and direction measured were averaged over 5 min, except for the maximum wind speed, which was recorded for each time interval. Important information about the long-range aerosol transport is derived from the analysis of the inverse trajectories of air masses that arrived in the region under study during the observations.

The concentrations of aerosol particles of 3 nm to 32 μm in size were recorded using a diffusion aerosol spectrometer [40], a photoelectric aerosol counter Grimm 1.109 (Grimm Aerosol Technik GmbH, Germany), and a five-stage Andersen-type impactor BB-35/25-4 (NII BP, Moscow, Russia) only at monitoring point L, as well as the concentrations of main gaseous pollutants, while air samples were taken at four points (Fig. 1). PM_{10} , $\text{PM}_{2.5}$, and PM_1 were calculated from the number aerosol concentrations by size and average density of particles. The latter was determined from the comparison between the simultaneous measurements with the Grimm 1.109 aerosol counter and the impactor. The CO_2 concentrations were measured with a Li-6262 gas analyzer (LICOR Inc., United States); CO concentrations, with Model 48C analyzer (Thermo Electron Corporation, United States); O_3 concentrations, with Model 49C analyzer (Thermo Electron Corporation, United States); NO_x concentrations, with a Model 200E chemical analyzer (Teledyne Instruments, United States); and SO_2 concentrations, with a Model 100E fluorescent analyzer (Teledyne Instruments, United States).



Fig. 1. Arrangement of sampling points and weather stations in the Gelendzhik region: L, P, and C mean Laboratory, Pier, and Cap, respectively; I means the station of the Institute of Oceanology, and V, the research vessel.

Atmospheric aerosols were sampled on AFA-CA-20 fiber filters [41] and then studied in the laboratory using the techniques recommended by domestic regulations [22, 30].

The elemental composition of the samples was determined by atomic absorption spectroscopy using the technique described in [22], C-115-M (Zapadpribor, Russia) and PerkinElmer 3110 (United States) spectrophotometers, and Russian State standard samples. The ranges of directly measured concentrations (in μg) of metals per 1 m^3 of pumped air are the following (the accuracy for the confidence probability $P = 0.95$ is given in parenthesis): 0.01–200 (12) for Cu, 0.01–100 (40) for Zn, 0.20–400 (27) for Fe, 0.02–400 (23) for Mn, 0.10–100 (25) for Al, 0.15–200 (31) for Ba, 0.04–10 (15) for Pb, 0.005–0.10 (10) for Cd, 0.05–2 (25) for As, 0.005–0.1 (22) for Ca, 0.008–0.16 (25) for Mg, 0.01–4 (30) for Na, 0.02–6 (25) for K, 0.01–200 (20) for Co, 0.05–2 (36) for Mo, 0.1–500 (31) for Cr, and 0.04–80 (23) for Ni.

The concentration of SO_4^{2-} ion in the soluble fraction was estimated by ion chromatography [42] at a TSVET-3006 instrument (OKBA NPO, Yoshkar-Ola, Russia). The method error is 25%; the detection threshold during pumping 1 m^3 air is $0.05 \mu\text{g}$.

The total protein concentration was determined by the fluorescence method using the dye described in [43]. The limits of the total protein relative error (with the confidence probability $P = 0.95$) of a Shimadzu RF-520 spectrometer (Shimadzu, Japan) were $\pm 25\%$ throughout the range of measurements; the range of

concentrations measured when pumping 1 m^3 of air was $0.003\text{--}50 \mu\text{g}$.

The concentration of polycyclic aromatic hydrocarbons (PAHs) was determined by high-performance liquid chromatography [44] using a Spectra Physics SP8800 (United States). The ranges of measured concentrations when pumping 1 m^3 of air are the following (μg): 0.005–78.2 for naphthalene, 0.006–66.2 for acenaphthylene, 0.005–18.2 for fluorene, 0.007–74 for acenaphthene, 0.005–11.8 for phenanthrene, 0.005–31.6 for anthracene, 0.005–36 for fluoranthene, 0.004–39.2 for pyrene, 0.005–15.2 for benzo(a)anthracene, 0.005–22.8 for benzo(b)fluoranthene, 0.004–20 for perylene, 0.001–27.4 for benzo(a)pyrene, 0.005–19.3 for 1,2,5,6-dibenzanthracene, and $0.003\text{--}16.6$ for benzo(ghi)perylene when sampling 9 m^3 of air. The error did not exceed 25%.

The concentration of total petroleum products (unsaturated hydrocarbons) was measured by IR Fourier-transform spectrometry [45] with a Varian FTS 1000 spectrometer (United States); the accuracy did not exceed 20%; the range of concentrations of petroleum products at pumping 1 m^3 of air was from 0.2 to 20 mg/m^3 .

Some biotoxins (for example, the toxin of blue-green algae often found in coastal areas [46], cholera toxin, and ricin) in atmospheric aerosol samples were detected by immunochromatography using the commercial kits SMART™ Cholera O1 (New Horizons Diagnostics Corporation, United States), SMART™ II Ricin (New Horizons Diagnostics Inc., United States), BioDetect™ Test Strips (Alexeter Technolo-

Table 1. Mass concentrations of aerosol fractions and certain aerosol components (Gelendzhik area, July 2009)

Date	PM ₁₀ , µg/m ³	PM _{2.5} , µg/m ³	PM ₁ , µg/m ³	Marine aerosol, µg/m ³	Total protein, µg/m ³
2	25.82 ± 4.38	14.15 ± 2.31	5.16 ± 0.78	1.11 ± 0.44	0.64 ± 0.35
3	17.57 ± 3.93	9.12 ± 2.48	4.38 ± 1.19	1.72 ± 0.96	0.95 ± 0.32
4	16.57 ± 3.68	9.88 ± 1.35	5.53 ± 0.66	0.89 ± 0.84	0.94 ± 0.08
5	14.87 ± 3.09	9.47 ± 1.96	5.09 ± 1.31	4.04 ± 1.60	1.52 ± 0.40
6	16.37 ± 4.76	9.73 ± 2.19	5.32 ± 1.31	1.29 ± 0.49	1.44 ± 0.53

gies, LLC, United States) or InstantChek Carbohydrate Identification Kit (EY Laboratories Inc., United States).

The concentration of viable microorganisms was determined by the standard culture method described in [47]. Filters with aerosol samples were poured with a nutrient medium containing 0.5% and 1.0% NaCl, then incubated for 6 h at 37°C, and plated at the same temperature to solid nutrient media with 0.5% and 1.0% of NaCl to obtain and count individual colonies (CFU).

Using the method of polymerase chain reaction and special primers and following sequencing by the method described in [47], the presence of genetic material of microorganisms and their taxonomic affiliation were detected.

Particles settled at various stages of the impactor were examined with a LEO1430VP scanning electron microscope equipped with an OXFORD energy spectrometer (Carl Zeiss, Germany), which allows surveying in the backscattered-electron mode to determine the elemental composition of individual particles (up to ~0.1 µm) or even their individual sections (for particles larger than 1 µm).

The weather station data were used to retrieve the 3D wind velocity field in the region under study using the algorithm described in [48]. Based on known wind speed and direction, the aerosol transport from a remote source was calculated and the concentration fields of controlled pollutants were constructed [49, 50]. The daily mean pollutant concentrations were calculated from the concentrations found. After that,

the pollutant concentration fields and API in the region were constructed.

RESULTS AND DISCUSSION

Figure 2 shows temperature, relative humidity, wind direction and speed, and air pressure measured during the study and averaged over 5-min intervals. The results show pronounced daily cycles of temperature, relative humidity, and solar radiation; alternating day and night wind directions, and a smooth change in air pressure.

During that period, the diurnal variations in the number concentrations of aerosol particles were more pronounced for nanometer particles. The daily mean PM₁₀, PM_{2.5}, and PM₁ concentrations (Table 1) were calculated from the Grimm 1.109 data with a mean particle density of (2.05 ± 0.14) g/cm³. The density of aerosol particles in the region under study was assessed from the comparison between the data of the aerosol particle counter and the impactor over six measurements. Table 1 shows that the mass of aerosol particles in the air of Gelendzhik was small: significantly smaller than the Russian standards [32] and smaller than in cities on the Mediterranean coast [3, 51–53] during the period of the experiment.

Airborne sounding of the atmosphere during the study made it possible to construct vertical profiles of some meteorological parameters and concentrations of pollutants. Clearly pronounced layers of high concentrations of CO, CO₂, O₃, and aerosol particles of 0.3–25 µm in size were observed at altitudes of up to 500 and up to 1000 m above sea level along the coast. No such layers were observed in the direction perpendicular to the coast. They were probably caused by the emissions of the nearest industrial center and port Novorossiysk and the corresponding wind direction during the airborne sounding.

APIs relating to gaseous pollutants and aerosol particles were calculated by Eq. (1) (Table 2). One can see that the concentrations of those substances during the observation period did not approach their MPC_{dm}, and the API did not exceed 0.84 for the sum of all the pollutants.

The results of chemical, elemental, and biological analyses of selected aerosol samples are presented in Tables 3–6. Atmospheric pollutants in low concentrations barely contribute to API; therefore, it is sufficient to determine the concentrations exceeding 1% of the

Table 2. Values of q_m/MPC_{dm} for gaseous pollutants and PM₁₀ in Gelendzhik in July 2009

Pollutant	MPC _{dm} , mg/m ³	Hazard category c_i	Date		
			3	5	6
CO ₂	9000	4	0.085	0.0850	0.0840
O ₃	0.16	1	0.450	0.3601	0.3280
CO	20	4	0.010	0.0130	0.0080
SO ₂	10	4	–	0.0006	0.0013
NO _x	5	4	–	0.0015	0.0018
PM ₁₀	0.06	3	0.293	0.2500	0.2600
API			0.838	0.7111	0.6831

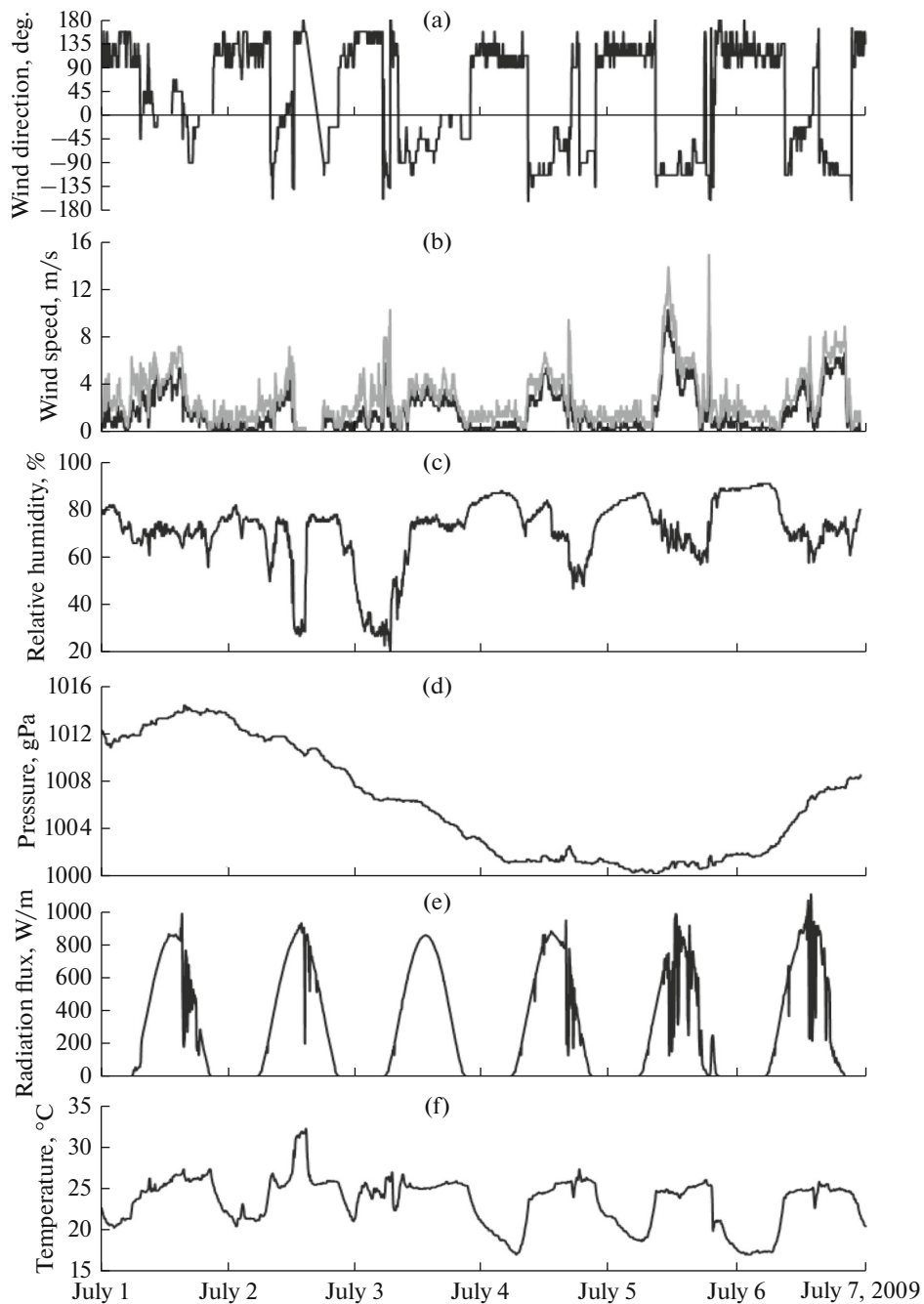


Fig. 2. Variations in the meteorological parameters in Gelendzhik during the study: (a) wind direction (western direction is taken as zero), (b) five-minute average wind speed (dark curve) and wind speed maximum over the period (light curve), (c) relative air humidity, (d) air pressure, (e) solar radiation, and (f) air temperature.

corresponding MPC. That is why no data are given in Tables 3–6 for those samples where the concentration of a substance to be determined does not exceed the detection threshold. In particular, PAHs and oil products did not exceed the detection threshold in any aerosol sample. The concentrations of As, B, Cd, Cr, Ni, and Ti are no higher than the detection thresholds in the samples taken at land points (see Table 3); and of As, B, Cd, Cr, Li, and Mo, in high-altitude samples (see Table 4). For all other substances, the detection

threshold was exceeded in at least one sample (see Tables 3–6), but in general their concentrations in aerosol turned out to be low. In high-altitude samples taken from onboard the aircraft during a flight along the coast at altitudes of 1000–2000 m in the vicinity of Gelendzhik, high concentrations of Ca, Ni, and Na in one sample were detected (see Table 4). They might be caused by the emission from Novorossiysk cement plants, which, as follows from the consideration of the inverse trajectories of air masses, could reach the area

Table 3. Concentrations of chemical elements and SO_4^{2-} ion ($\mu\text{g}/\text{m}^3$) in atmospheric air samples taken at different land points in Gelendzhik (gray cells correspond to the laboratory sampling, from 10:00 on July 7 to 10:00 on July 6, 2009)

Pollutant	Date																				
	July 2				July 3				July 4				July 5	July 5–6							
	V	P	C	L	V	P	C	L	V	P	C	L	V	10–13	13–16	16–19	19–22	22–01	01–04	04–07	07–10
Al	0.073	0.081	0.111	0.215	0.143	0.080	0.016	0.051	0.047	0.038	0.039	0.099	0.073	0.074	0.095	0.068	0.066	0.046	–	0.074	0.065
Ba	0.007	0.004	0.007	0.012	0.012	0.002	0.001	0.004	0.004	–	0.001	0.008	0.003	0.003	0.000	0.004	0.009	0.009	0.004	0.012	0.015
Ca	2.403	4.344	8.960	6.060	5.305	1.705	1.855	5.072	2.833	1.428	2.055	5.960	1.628	3.400	3.044	2.772	3.022	4.622	1.055	4.166	4.944
Co	–	–	–	–	–	–	–	–	0.012	0.011	0.014	0.011	0.009	0.012	0.012	0.013	0.011	0.012	0.013	0.014	0.011
Cu	0.003	–	0.320	0.128	0.003	0.220	–	–	–	–	0.068	0.028	–	0.108	0.372	–	–	0.114	0.006	0.004	0.444
Fe	–	0.044	0.100	0.344	–	–	–	–	–	0.078	0.044	0.144	–	0.111	–	–	–	–	–	–	–
K	0.154	0.102	0.375	0.310	0.281	0.084	0.107	0.134	0.228	0.100	0.202	0.139	0.145	0.287	0.280	0.163	0.255	0.073	0.009	0.231	0.305
Li	–	–	0.003	0.006	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	0.003
Mg	0.370	0.490	0.460	0.524	0.620	0.177	0.152	0.172	0.273	0.174	0.153	0.173	0.341	0.276	0.285	0.103	0.012	–	–	–	0.100
Mn	0.014	0.010	0.025	0.034	0.024	0.005	0.003	0.015	–	0.006	0.007	0.024	–	0.010	0.006	0.007	0.009	0.009	0.001	0.003	0.010
Mo	–	–	–	–	–	–	–	–	–	–	–	–	–	–	0.024	0.031	–	0.042	–	–	0.034
Na	0.508	0.636	0.234	0.360	1.202	0.524	0.323	0.658	0.824	0.347	0.147	0.092	2.446	1.558	1.758	1.480	0.691	0.227	0.636	0.536	0.624
P	–	–	0.349	0.120	<0.04	–	–	–	–	0.049	0.097	0.118	–	0.105	0.075	–	–	–	–	–	0.079
Pb	0.070	0.084	–	0.015	0.128	0.002	0.306	0.139	0.105	0.173	–	0.029	–	–	–	0.223	0.177	–	0.313	–	0.032
SO_4^{2-}	2.879	3.444	3.777	4.999	5.799	3.733	3.233	3.233	2.066	3.999	2.444	3.999	3.888	4.999	3.999	5.333	3.000	2.666	2.333	2.333	3.666
Si	0.125	0.092	0.136	0.409	0.235	–	–	–	0.042	–	–	0.035	0.170	–	–	–	–	–	–	0.021	0.036
Sr	0.024	0.047	0.063	0.059	0.058	0.014	0.015	0.027	0.016	0.012	0.013	0.030	0.014	0.022	0.018	0.018	0.012	0.017	0.005	0.013	0.019
Ti	<0.005	<0.005	0.006	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Zn	0.046	0.036	0.090	0.100	0.083	0.051	0.013	0.025	0.300	–	–	0.006	0.063	0.012	–	0.008	–	0.061	–	0.007	0.038
Σ	6.676	9.414	15.016	13.695	13.897	6.597	6.024	9.530	6.749	6.414	5.284	10.895	8.775	10.977	9.968	10.227	7.267	7.898	4.371	7.414	10.431

“–” means concentrations below the detection threshold; for Mo, $0.01\text{MPC}_{\text{dm}} = 5 \mu\text{g}/\text{m}^3$ [31].

Table 4. Concentrations of chemical elements and SO₄²⁻ ion (µg/m³) in atmospheric air samples taken during aircraft sounding in Gelendzhik in July 2009

Pollutant	Date, time, sampling altitude, flight direction					MPC [31]
	July 5, 15:23–15:55, 2100 m, along the coast toward Anapa	July 6, 13:03–13:18, 2000 m, along the coast toward Sochi	July 6, 13:52–14:05, 1500 m, along the coast toward Anapa	July 6, 14:07–14:20, 1000 m, along the coast toward Sochi	July 6, 14:44–15:12, 500 m, along the coast toward Anapa	
Al	0.126	0.166	0.906	0.340	0.194	20
Ba	0.010	0.017	0.066	0.028	0.017	15
Ca	3.880	6.017	17.488	10.047	4.465	10
Co	–	0.012	0.014	0.011	0.007	0.1
Cu	0.013	0.008	0.041	0.028	0.012	10
Fe	0.254	0.473	3.115	0.719	0.341	100
K	0.097	0.104	0.483	0.272	0.321	10
Mg	0.650	1.077	1.790	2.488	0.692	100
Mn	0.011	0.012	0.059	0.034	0.021	10
Na	0.802	1.357	2.204	1.913	1.255	2
Ni	0.034	0.128	0.063	0.083	0.037	0.05
P	0.037	0.112	0.173	0.260	0.175	0.3
Pb	0.049	–	–	0.198	0.097	0.5
SO ₄ ²⁻	4.140	4.544	8.785	8.508	4.319	–
Si	0.137	0.170	1.717	0.430	0.221	5
Sr	0.037	0.054	0.095	0.082	0.036	5
Ti	0.004	0.008	0.032	0.011	0.005	10
Zn	0.049	0.075	0.287	0.108	0.053	1
Σ	10.33	14.33	37.35	25.56	12.27	

Cells with values above MPC for residential areas are gray.

Table 5. Concentrations of total protein (ng/m³) in atmospheric air samples taken at different land points in Gelendzhik in July 2009

Date and time of sampling	Sampling point			
	V	P	C	L
July 2, 10:00–13:00	218	503	957	894
July 3, 10:00–13:00	1402	885	841	672
July 4, 10:00–13:00	903	1063	934	876
July 5, 10:00–13:00	1063	N.d.	N.d.	1618
July 5, 13:00–16:00	N.d.	N.d.	N.d.	585
July 5, 16:00–19:00	N.d.	N.d.	N.d.	1189
July 5, 19:00–22:00	N.d.	N.d.	N.d.	1369
July 5, 22:00–01:00	N.d.	N.d.	N.d.	1659
July 6, 01:00–04:00	N.d.	N.d.	N.d.	2361
July 6, 04:00–07:00	N.d.	N.d.	N.d.	1670
July 6, 07:00–10:00	N.d.	N.d.	N.d.	1148
July 6, 10:00–13:00	N.d.	894	1090	1455

“N.d.” means absence of data.

surveyed during the period of airborne sounding. The total mass of aerosol particles, defined as the sum of the masses of the controlled chemical elements and ions, exceeded 37 µg/m³ at the maximum, while it was always lower than 15 µg/m³ in land-taken samples. This fact confirms our assumption that, most likely, the polluted air masses arrived in the region under study from the neighboring region.

Having data on the chemical composition of the aerosol, information about its possible sources can be derived. Marine aerosol is detected by the presence of Na in aerosol particles, since, as shown in [54], Cl₂ molecules can form in the atmosphere due to surface reactions, which leave the particles thus disturbing the balance between the Na and Cl concentrations in the saline aerosol. The concentration of biogenic aerosol

Table 6. Concentrations of total protein (ng/m^3) in atmospheric air samples taken during aircraft sounding in Gelendzhik in July 2009

Date, time, sampling altitude, flight direction	July 5, 14:33–15:00, 2100 m, along the coast toward Sochi	July 6, 12:46–13:01, 2000 m, perpendicular to the coast	July 6, 13:36–13:49, 1500 m, perpendicular to the coast	July 6, 13:36–13:49, 1000 m, perpendicular to the coast	July 6, 14:44–15:12, 500 m, along the coast toward Anapa
Concentration	415	508	452	434	521

Table 7. API (defined as the sum of five maximal values) and its main components for different substances entering aerosol particles and PM_{10} at point L in Gelendzhik in July 2009

Pollutant	$\text{MPC}_{\text{dm}}, \mu\text{g}/\text{m}^3$	Hazard category c_i	Date of measurements			
			2	3	4	5
Al	30	2/1.3	0.00080	0.00040	0.00030	0.00070
Ba	4	1/1.7	0.00003	0.00001	0.00001	0.00003
Ca	20	2/1.3	0.12800	0.08600	0.09400	0.17400
Co	0.4	2/1.3	—	—	0.00780	0.01900
Cu	3	2/1.3	—	—	—	0.03450
K	50	2/1.3	0.0008	0.00060	0.00060	0.00150
Mg	1	2/1.3	0.2380	0.29300	0.12000	0.00290
Mn	50	3/1.0	0.0004	0.00030	0.00020	—
Na	100	3/1.0	0.0021	0.00680	0.00460	0.01750
SO_4^{2-}	20	3/1.0	0.1440	0.15900	0.11200	0.29900
Zn	50	2/1.3	—	—	—	0.00005
Protein	100	2/1.3	0.0011	0.00190	0.00170	0.00610
PM_{10}	60			0.29300		0.25000
API (without PM_{10})			0.5132	0.54670	0.33840	0.53110

is determined by the total protein; the emissions of automobile and sea transport, by the concentrations of PAHs, petroleum products, and some other substances. Other sources can also contribute to the aerosol concentration.

The analysis of data on the aerosol chemical composition and mass showed that marine aerosol amounted 4 to 25% of PM_{10} mass during the observation period (Table 1). The microscopic analysis of the chemical composition of particles deposited at different impactor stages showed that the percentage of marine aerosol was 3–10% of aerosol particles larger than $0.1 \mu\text{m}$. The analysis accuracy is low, since the procedure for gathering information on the chemical composition of a large number of individual particles is laborious. It is difficult to determine the volume of particles of complex shapes; many particles have a heterogeneous chemical composition, etc. For the same reasons, it is extremely difficult to obtain similar information for different particle size fractions. The technique makes it possible to detect only relatively large particles; therefore, it is impossible to study particles smaller than $\sim 0.1 \mu\text{m}$ and absorption layers of organic and bioorganic compounds on the surface of larger particles. However, this technique provides unique

information about individual particles within atmospheric aerosol.

The analysis of particles found out at different stages of the impactor, which operated under the prevailing off-shore wind, showed that there were few particles of marine origin (in such particles, Na, Cl, Mn, Mg, S, and O should be present in large amounts). The fraction of marine particles in the samples taken under different wind directions is small (Table 1), which well agrees with data of other authors [51–54]. The same indirectly follows from work [55], where a small decrease in the aerosol concentration has been noted when the wind is changing from off-shore to on-shore. The percentage of marine aerosol attains 20% in summer in the Eastern Mediterranean [33], which is a little higher than the microscopically detected values; and near Rome, 3–7% on the average in October–July. Let us emphasize that total aerosol, detected in these studies, is much higher.

Among particles found out at different impactor stages, 60–80% of particles are of soil origin (with O, Si, and/or Al in large amounts) or, probably, anthropogenic. Particles with a large content of Pb are related, most probably, to emissions of vehicles, where fuel with Pb additives is used, and to road dust. Note that the analysis of large particles (tens of microme-

ters) showed inhomogeneous particle composition over the volume. The same is witnessed by the presence of carbon (related, probably, to surface-absorbed organic compounds [34, 36, 37, 57, 58] within soil and anthropogenic particles, including aerosols formed during cooking.

The most difficult is to detect the mass of biogenic components of atmospheric aerosol from microscopic analysis data, since not only individual biogenic particles, but also particle-surface adsorbed layers contribute to it. The chemical analysis showed (see Tables 5 and 6) that the total protein was from 0.22 to 2.4 $\mu\text{g}/\text{m}^3$ in the aerosol samples under study, which was from 2.5 to 10% of the total aerosol, according to Table 1. These values are comparable with literature data on the content of biogenic components in atmospheric aerosol [59, 60]. During the sampling period, the total protein in the surface air layer near Gelendzhik exceeded that at altitudes of 1000–2000 m, which was evidence of the prevailing contribution of local sources of bioaerosol as compared to distant sources. The daily variations in the total protein in the surface air layer show a night maximum (see Table 3). Therefore, since off-shore wind was usual during nighttime observations (night breeze, see Fig. 2a), one can conclude that main sources of protein were located on land, not in the sea. Note that the situation is the reverse for Na and SO_4 (see Table 3), that is, during the day breeze from the sea, their concentrations in atmospheric aerosol are maximal, which is evidence of primarily marine origin of these components.

Since neither PAHs nor oil products were detected in the aerosol samples within the limits of technique sensitivities, the contribution of vehicles in aerosol pollution is small.

The concentration of viable microorganisms in atmospheric aerosol is also small in the region. Viable microorganisms with the mean concentration ~ 2000 CFU/ m^3 were detected in 75% of atmospheric air samples taken at land and over-water sampling points; the concentrations differed by up to 100 times in different samples. These values are typical for other regions [47, 57–62] but are not dangerous for people if these microorganisms are not pathogenic. The genetic analysis of grown colonies of microorganisms has not shown human disease-producing germs among about 40 isolates identified (*Bacillus cereus* (S1 strain), *Bacillus megaterium* (CCM14B and M1Pca strains), *Bacillus subtilis* (DSM 6405 strain), *Bacillus anthracis* (HCYL02 strain), etc.)). The more detailed analysis of microorganisms in regional aerosol was beyond the scope of our study. The $q_m/\text{MPC}_{\text{dm}}$ values are low for microorganisms; therefore, they are not included in API indices.

Based on the experimentally detected concentrations of aerosol pollutant, APIs were calculated by Eq. (1). For comparing API values found from different numbers of pollutant concentrations, only the five highest $q_m/\text{MPC}_{\text{dm}}$ values were included in the index

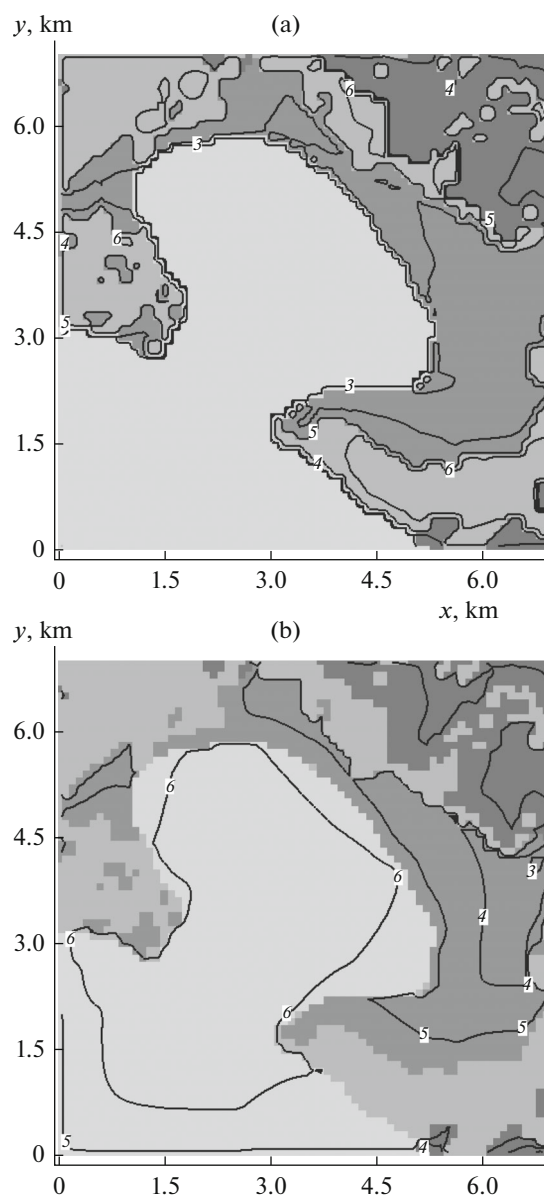


Fig. 3. Total protein concentration fields (ng/m^3) on (a) July 2 from 10:00 to 13:00 (isolines 10.0 (3), 100.0 (4), 1000.0 (5), and 2500.0 (6)) and (b) July 3, 2009, from 10:00 to 13:00 (isolines 1.0 (3); 10.0 (4); 100.0 (5), and 1000.0 (6)): water surface (□), vegetation (▨), buildings (▩), and free space (■).

for point L (Table 7). Jointly considering data in Tables 2 and 7, one can easily note that the API calculated for the sum of gaseous and aerosol pollutants did not exceed unity throughout the observation period. No one $q_m/\text{MPC}_{\text{dm}}$ value approaches unity. Gaseous pollutants and the total aerosol mainly contributed in the APIs in that period. According to Table 2, $q_m/\text{MPC}_{\text{dm}} \leq 0.3$ for PM_{10} , while the sum of $q_m/\text{MPC}_{\text{dm}}$ for all the controllable pollutants is about two times higher (see Table 7). In our opinion, this

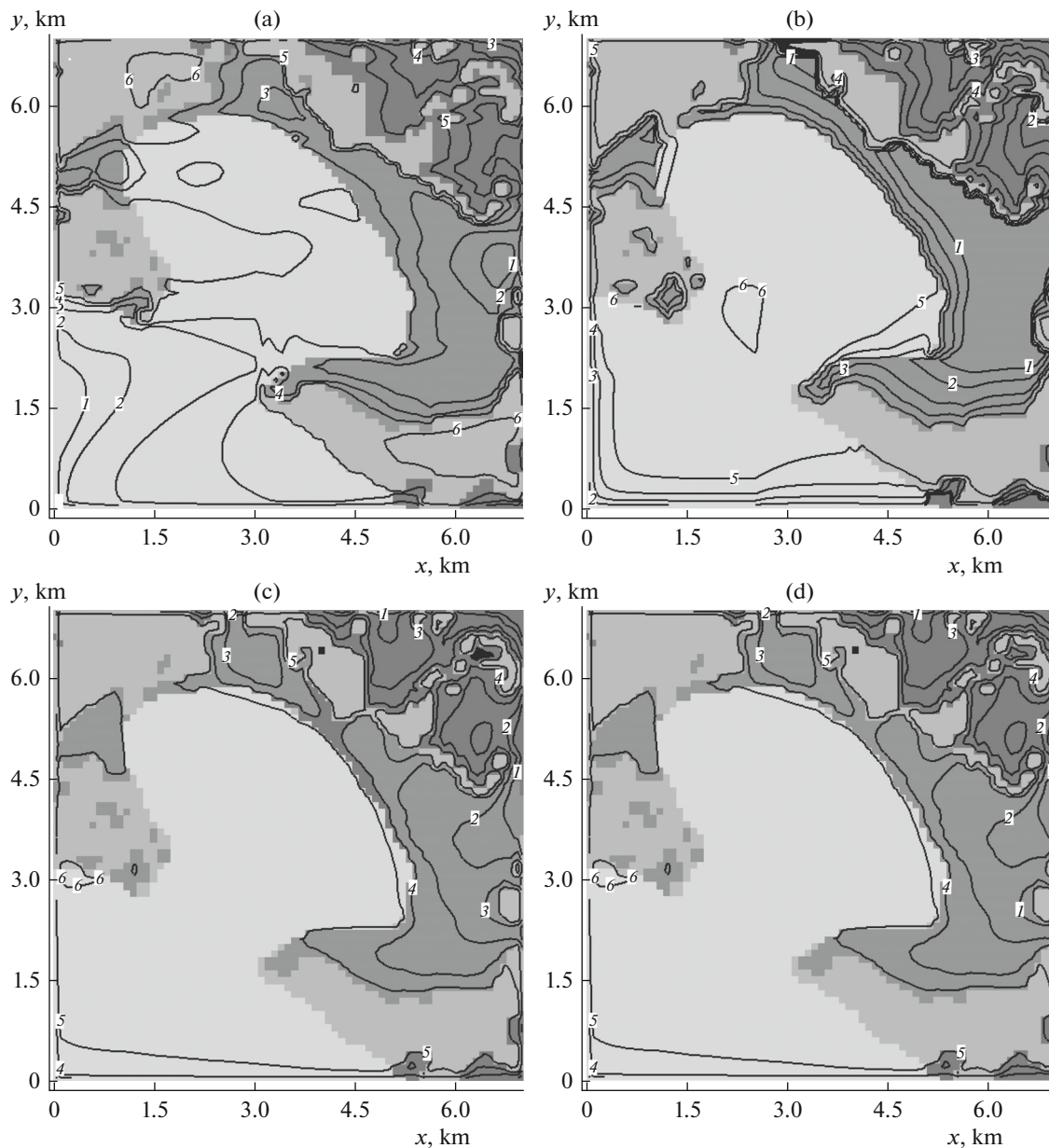


Fig. 4. API fields for (a) July 2 (isolines 0.01 (1), 0.02 (2), 0.05 (3), 0.1 (4), 0.2 (5), and 0.6 (6)), (b) July 3 (isolines 0.01 (1), 0.02 (2), 0.05 (3), 0.1 (4), 0.2 (5), and 0.6 (6)), (c) July 4 (isolines 0.01 (1), 0.02 (2), 0.05 (3), 0.1 (4), 0.2 (5), and 0.4 (6)), and (d) July 5, 2009 (isolines 0.01 (1), 0.02 (2), 0.05 (3), 0.1 (4), 0.2 (5), and 0.5 (6)): water surface (■), vegetation (■), buildings (■), and free space (■).

proves that MPC_{dm} for PM_{10} is highly overestimated in Russian regulations.

In an ideal case, q_m/MPC_{dm} for PM_{10} and the sum of q_m/MPC_{dm} for all the controllable pollutants should be equal. However, when changing the chemical composition of aerosol particles, the sum of q_m/MPC_{dm} for all pollutants detectable in aerosol changes. Hence, it is evident that measurements of q_m/MPC_{dm} for all the controllable aerosol fractions are of higher information content than q_m/MPC_{dm} for PM_{10} (which is easier and cheaper to measure); therefore, the former can be

advised for a more accurate assessment of the atmospheric air quality.

Following the above described technique, fields of regional pollutions can be constructed for aerosol pollutants measured at several points. Figure 3 shows fields of regional pollution with total protein included in the composition of the aerosol. It is seen that the main sources of total protein were on land on July 2, 2009, where its concentration was much higher than over the water, while the situation was the reverse on July 3, 2009. This means that there are sources of bio-aerosols of different natures in the region, and their

powers are not constant. Similar and even more detailed conclusions can be drawn from temporal variations in the pollution fields constructed for other substances.

After constructing regional pollution fields for individual substances and averaging their values to assess their daily mean concentrations, $q_m/\text{MPC}_{\text{dm}}$ can be calculated for all pollutants detectable in aerosol and the API for the region under study for every day of observations. Figure 4 exemplifies isolines of the API field for pollutants in the aerosol for the territory and water area of Gelendzhik.

Figure 4 proves that API (air quality) strongly depends on the observation site and can change by several times. The region of maximal pollution drifted in the region versus meteorological conditions, though API maxima for aerosol pollutants weakly changed during the observation period. All this shows a need for measuring atmospheric pollution simultaneously at several points to map the regional air quality with high information content.

CONCLUSIONS

The study has shown that the complex assessment of atmospheric air quality is a complicated problem. Even the short-term experiment in Gelendzhik required large resources: several land (water) sampling points, aircraft sounding of the atmosphere, a variety of measuring and sampling instruments, different analysis techniques, and mathematical simulation of aerosol propagation in the surface air layer.

Estimation of concentrations of different gaseous and aerosol pollutants allowed calculation of the complex index of atmospheric pollution at a measurement point and construction of fields for changing this index in the region. It is shown that both API and its regional distribution is strongly affected by meteorological processes. The assumed correlation between the atmospheric concentrations of different pollutants and their sources allows us to attempt prediction of the API by the weather conditions forecasted and anthropogenic activity in the future.

During the period of our study, APIs were low both for the complex of gaseous pollutants and atmospheric aerosol and for the complex of pollutants within aerosol particles. No pollutants in concentrations above MPC were detected in that period. We conclude that Gelendzhik was an ecologically clean city with high-quality atmospheric air during the period under study.

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