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ESTIMATION OF THE GROUND-LEVEL OZONE LIFETIME UNDER RURAL CONDITIONS

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The investigation of the ozone formation and destruction is of great interest because ozone influences the atmospheric chemistry and plays an important role in the climate change. The ground-level ozone lifetime alters depending on physical and chemical properties of the locality, meteorological factors, atmospheric turbulence, and other conditions. In this paper, the method of estimation of the ozone lifetime under rural conditions is presented. The discrepancy between the solar intensity as well as related turbulent air mixing maximum and the ozone concentration maximum during the day was used in the lifetime calculations. The solar radiation intensity and duration were taken as main parameters in calculating the ozone lifetime by estimating the ozone enrichment at the ground level. In the calculations the least-squares method was applied. The ground-level average ozone lifetime was estimated to be in the range of 3.6–5.6 hours at the rural Preila station and in the range of 2.8–3.9 hours at the Rūgšteliškis station during different seasons.

Keywords: ozone lifetime, solar radiation intensity and duration, seasons, rural conditions

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1. Introduction

The ground-level ozone due to its reactivity is an important minor gaseous constituent of the troposphere. Ozone and related atmospheric oxidants play a significant role in controlling the chemical lifetimes and reaction products of many atmospheric species and also influence the organic aerosol formation [1]. The two major sources of natural ground-level ozone can be emphasized: hydrocarbons released by plants and soil, as well as small amounts of stratospheric ozone, which occasionally migrate down to the Earth's surface [1]. A significant part of ozone of anthropogenic origin is the outcome of photochemical reactions between oxides of nitrogen (NO_x) and volatile organic compounds (VOCs) emitted from the anthropogenic sources in the presence of sunlight. The main ozone source in the remote locations is not local anthropogenic photochemical reactions but the ozone transport from the polluted regions [2, 3]. Thus, transport of ozone from other locations is more important than the local ozone production in the majority of rural regions. The ozone transport is often evaluated by using various regional transport models requiring data of the ozone lifetime. However, particular ozone lifetimes are difficult to characterize, since ozone is a secondary species resulting from a complex series of chemical reactions, for example, the lifetime of tropospheric ozone is considered to be of the order of 1 to 2 days in summer [4]. The data on the ozone lifetime in the atmosphere show that it can depend on the air pollution level, meteorological conditions, etc. Furthermore, the ozone lifetime varies according to latitude and altitude, and also can vary over years. According to [5], the ozone lifetime has decreased by approximately 30% since 1890, especially after 1930.

The dry deposition of ozone is one of the most important sinks in the boundary layer ozone budget [6]. The latest investigations [7] showed that the changes in dry deposition to vegetation (not including changes in aerodynamic resistance) could account for up to 80% of the surface ozone change in Spain. The dry deposition rate of the ground-level ozone is particularly dependent on the underlying surface features. The experiments [8] showed that the deposition rate to the sea surface varied in the interval of 0.53–1.1 mm s^{-1}, whereas the maximum ozone deposition rate for pastures [9] could reach 7 mm s^{-1}, which is about threefold lower than values derived for forests. Thereby, the nature of underlying surface can determine the ground-level ozone lifetime. Therefore, the ozone lifetime is not steady and depends

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not only on the pollutant concentration in the air but also on the vertical or horizontal pollutant transport intensity that determines the ozone contact time with underlying surface. Mostly, the vertical mixing is strong in the boundary layer and ozone levels reach maximum values in the afternoon due to local photochemical formation and vertical transport from aloft to the underlying surface [10]. As the land surface is heated by solar energy and convection is initiated, the ozone-rich air is entrained from the upper layers in the troposphere into the boundary layer and is mixed down to the surface. Stevenson et al. [11] estimated the lifetime of tropospheric ozone to be about 22 days, whereas according to [12, 13] the ozone lifetime is only a few days in the continental boundary layer in summer but several weeks in the free troposphere. The tropospheric ozone lifetime in the report of Intergovernmental Panel on Climate Change [14] is estimated to be varying from some hours to some days. All available data suggest that there are plenty of uncertainties in assessing the ozone lifetime under different conditions.

The aim of this work was to develop a method for estimation of the ground-level ozone lifetime under rural conditions during different seasons.

2. Experiment

The ground-level ozone concentration and ultraviolet (UV) radiation measurements were performed at two rural stations in Lithuania. The Preila station (55°22′ N and 21°02′ E of 5 m above the sea level) is located in western Lithuania on a coast of the Baltic Sea, on the Curonian Spit. The other rural station Rūgšteliškis (55°27′ N and 26°00′ E, 170 m a. s. l.) is located in the eastern part of Lithuania in a forested, hilly area at about 350 km distance from the Baltic Sea. The ozone sampling height at both stations was 2.5 m above the ground. There are no large local sources of anthropogenic pollution close to the monitoring sites. The ozone concentrations were measured with the commercial UV absorption ozone analyzers O341M (*Environnement s.a.*) at the stations in Preila and ML9811 (*Monitoring Labs*) in Rūgšteliškis. The ozone detection limit was 2 μg m^{-3}.

The UVB radiation at the stations was measured with the pyranometer SKU–430 (*Skye Instrument Ltd.*) with a measurement range of 0–5 W m^{-2}, providing radiation registration in the intervals of 280–315 nm. The sensor sensitivity is 150 mV W^{-1} m^2. Sensors of the solar radiation are installed on the top of the meteorological tower at the height of 26 m above the ground level in Rūgšteliškis and at the 10 m height in Preila. All measurements proceeded in a continuous mode with hourly data resolution. The data of the cloudless days were used in the analysis. The period of two years, 2004 and 2005, was chosen for the analysis. The mixing heights were taken from the National Oceanic and Atmospheric Administration (NOAA) Air Resources laboratory (ARL) Real-time Environmental Applications and Display sYstem (READY) website using HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) model [15].

3. Modelling

The earlier investigations [16] have shown that the main source of the ground-level ozone concentration at the Preila station is the transport of ozone from other locations but not the local photochemical production. Therefore, in our study we have assumed that the ozone lifetime in the environment of this station is mainly determined by the ozone transport from higher layers to the ground-level through air turbulence, and destruction at the Earth's surface but not by the chemical loss. Data were grouped into the four clusters – seasons (winter, spring, summer, and autumn) chosen in the work taking into account a different ozone concentration level, ozone formation peculiarities, underlying surface conditions, etc. As mentioned above, the dry deposition of ozone to the underlying surfaces is regulated by atmospheric turbulence. The main reason for atmospheric turbulence, however, is incoming solar radiation, the intensity of which varies during the year. The data of hourly UVB radiation intensity were used to determine its average diurnal courses during separate seasons (Fig. 1(a)). UV radiation showed the same diurnal pattern as the total solar radiation intensity during cloudless days: the maximum of global and UV radiation intensities is observed at the same local time (Fig. 1(a)) and only magnitudes differ by approximately a constant factor. The diurnal courses of the mixing heights were determined for the same days and periods (Fig. 1(b)) as the solar radiation. By noon, the mixing height typically exceeds 1000 m and ozone is well mixed within the mixing layer [17]. A close relationship exists between the surface temperature and the solar radiation intensity, while surface temperature changes have a strong effect on the mixing height h [18]. That is clearly evident from the diurnal courses of the mixing height (Fig. 1(b)): the maxima of the magnitudes of the solar radiation intensity and the mixing height were observed at the same time.

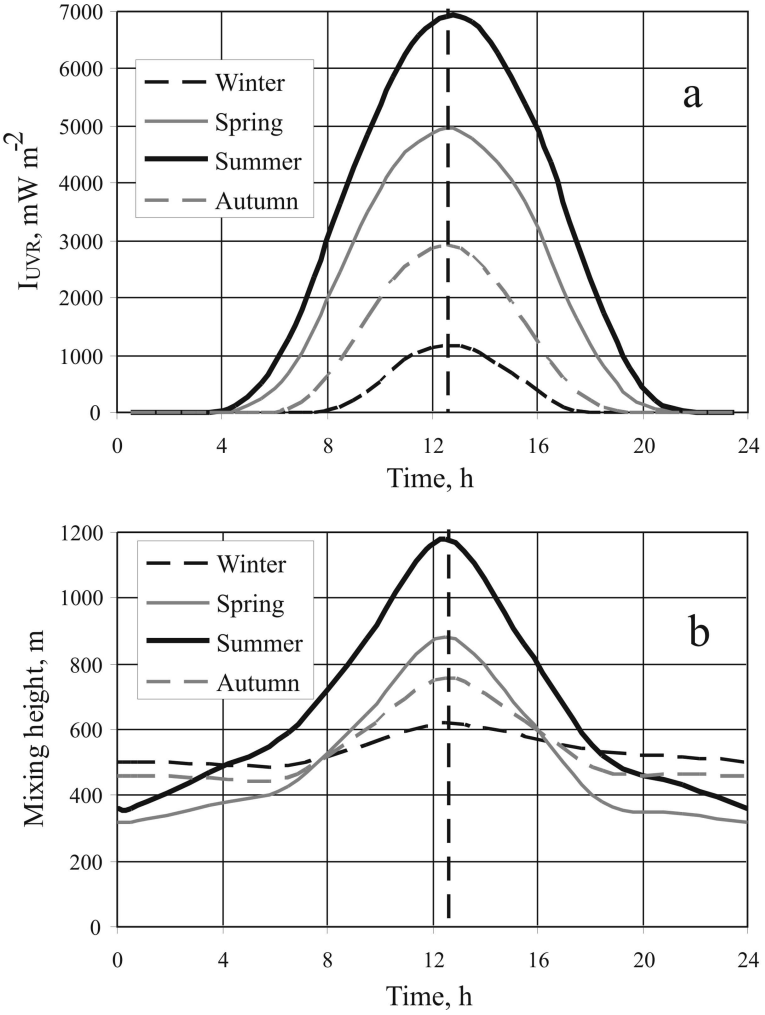

Fig. 1. Diurnal courses of (a) the average solar radiation intensity and (b) the average mixing height at the Preila station.

The course of the solar radiation intensity can be described by Eq. (1) as a function of time from the sunrise:

$$I = I_0 \frac{1 - \cos\left(\frac{2\pi}{\tau_\mathrm{s}} t\right)}{2}\,, \tag{1}$$

where τ_s is the time from sunrise to sunset, I_0 is the maximum magnitude of solar radiation, t is the day time.

It was supposed that the air enrichment with ozone due to the turbulent mixing is proportional to the solar radiation intensity near the Earth's surface. The stable nocturnal boundary layer and a very low ozone level in the station surroundings are observed rarely [19]. The low turbulence intensity is expected at night; therefore, the air enrichment with ozone ($\mu\mathrm{g\,m^{-3}h^{-1}}$) from the upper layers due to the turbulent mixing in the atmosphere can be expressed as

$$E = E_0 + E_\mathrm{S} \frac{1 - \cos(\omega t)}{2}\,, \tag{2}$$

with

$$\omega = \frac{2\pi}{\tau_\mathrm{s}}\,, \tag{3}$$

where E_0 is the air enrichment with ozone during the night hours, E_S is the air enrichment under maximum turbulent mixing conditions in the atmosphere, ω is the cyclic frequency.

The sunrise time is considered to be $t = 0$, so experimental data are moved on the time scale proportionally. Then, the course of the ozone concentration c from the sunrise can be described as a differential equation:

$$\frac{\mathrm{d}c}{\mathrm{d}t} = -\lambda c + E_0 + \frac{E_\mathrm{S}}{2}\left(1 - \cos\omega t\right)\,, \tag{4}$$

where λ is the ozone decay rate.

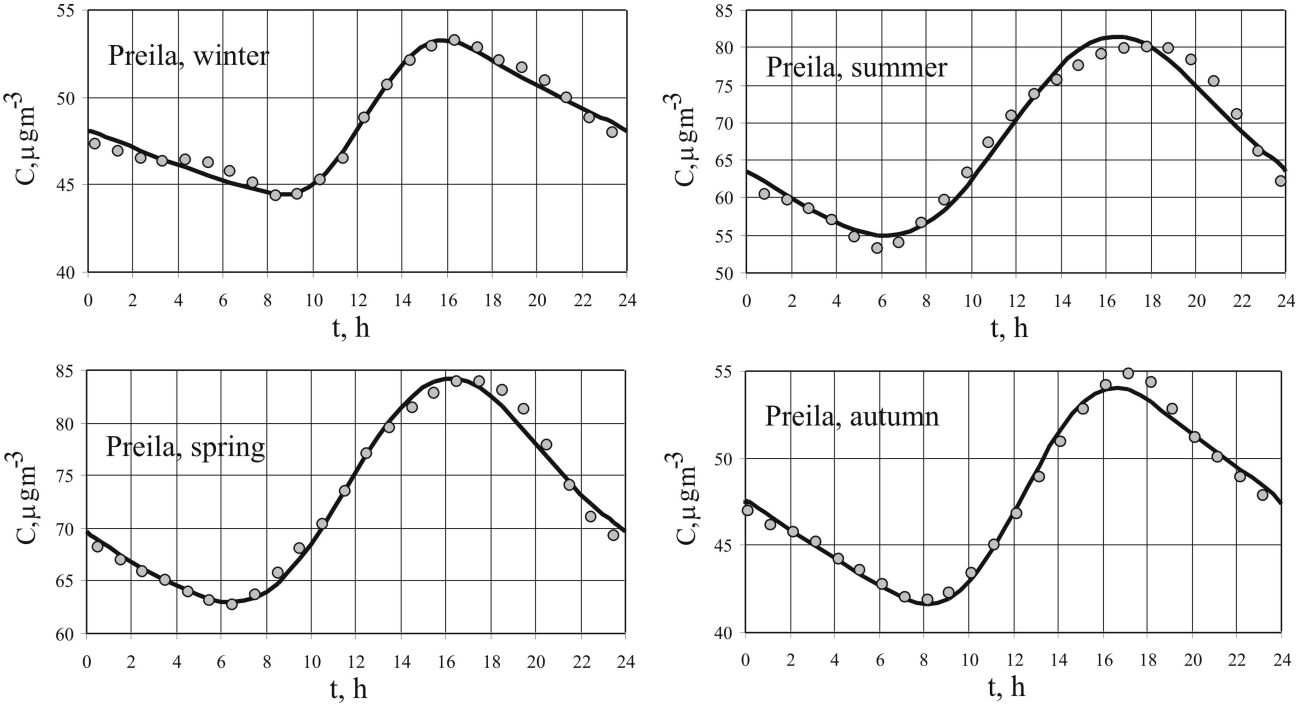

Fig. 2. Diurnal courses of the observed (dots) and calculated (solid line) ozone concentrations at the Preila station during different seasons.

When $t < \tau_{\rm s}$, the solution of the equation is

$$c = \frac{E_0}{\lambda} + E_{\rm S}\left[\frac{1}{2\lambda} - \frac{\lambda\cos\omega t + \omega\sin\omega t}{2(\lambda^2+\omega^2)} - \frac{\omega^2\left[1-{\rm e}^{\lambda(\tau_{\rm s}-T)}\right]}{2\lambda(\lambda^2+\omega^2)(1-{\rm e}^{-\lambda T})}{\rm e}^{-\lambda t}\right], \quad (5)$$

and when $\tau_{\rm s} < t < T$, where T is 24 hours, the solution is

$$c = \frac{E_0}{\lambda} + E_{\rm S}\frac{\omega^2\left[1-{\rm e}^{-\lambda\tau_{\rm s}}\right]}{2\lambda\left(\lambda^2+\omega^2\right)\left(1-{\rm e}^{-\lambda T}\right)}{\rm e}^{\lambda(\tau_{\rm s}-t)}\,. \quad (6)$$

The least-squares method is applied in most simulation tasks, in which the experimental data are used. The least-squares method was applied to determine parameters E_0, $E_{\rm S}$, and λ. The sum S of experimental and theoretical values of squares of differences is

$$S = \sum_{i=1}^{n}\left(c_i - E_0\frac{1}{\lambda} - E_{\rm S}\varphi_i\right)^2, \quad (7)$$

where c_i is the experimental ozone concentration, n is the number of experimental values, φ_i is the multiplier factor of $E_{\rm S}$ in Eq. (5) when $t_i < \tau_{\rm s}$, and in Eq. (6) when $\tau_{\rm s} < t_i < T$.

Assuming that $\partial S/\partial E_0 = 0$ and $\partial S/\partial E_{\rm S} = 0$, the equation system will be

$$\begin{cases} E_0\dfrac{n}{\lambda^2} + E_{\rm S}\displaystyle\sum_{i=1}^{n}\frac{\varphi_i}{\lambda} = \sum_{i=1}^{n}\frac{c_i}{\lambda}\,, \\ E_0\displaystyle\sum_{i=1}^{n}\frac{\varphi_i}{\lambda} + E_{\rm S}\sum_{i=1}^{n}\varphi_i^2 = \sum_{i=1}^{n}c_i\varphi_i\,. \end{cases} \quad (8)$$

E_0 and $E_{\rm S}$ values will be found by solving equation system (8) for the definite parameter λ.

By changing the parameter λ, the minimum of the sum S is determined and values of λ, E_0, and $E_{\rm S}$ are found. The average ozone lifetime τ can be expressed as

$$\tau = \frac{1}{\lambda}\,. \quad (9)$$

The initial day time in the graphic representation was restored by adding the sunrise time.

4. Results and discussion

The diurnal courses of the observed and calculated average ozone concentrations during the four seasons are shown in Fig. 2. A good coincidence of experimental and calculated data indicates that the model can be used for the evaluation of the ground-level ozone lifetime under the rural coastal conditions.

The same model was applied by using data from the other inland station R$\bar{\rm u}$gšteliškis. The simulation

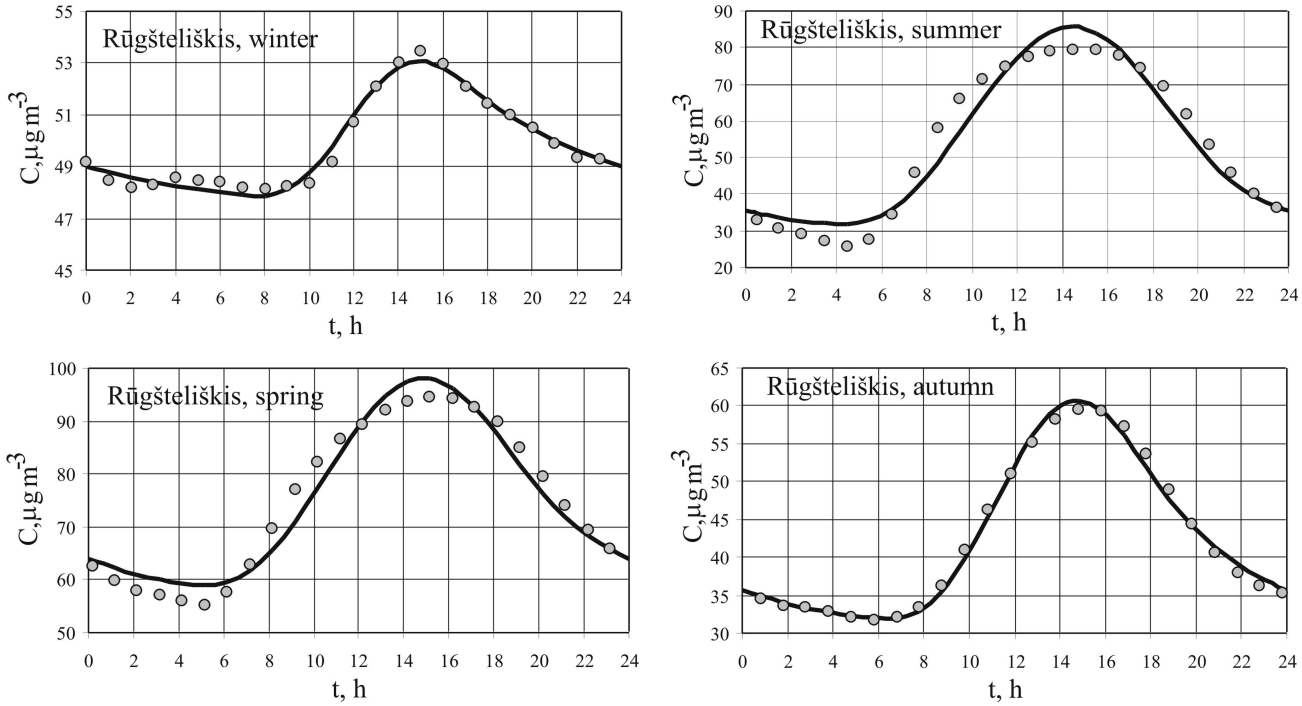

Fig. 3. Diurnal courses of the observed (dots) and calculated (solid line) ozone concentrations at the Rūgšteliškis station during different seasons.

results show the enhanced calculated ozone values in comparison with the data observed during early morning and midday hours in spring and summer but the discrepancy is only about 10 and 20% during midday and early morning hours, respectively (Fig. 3). One of the reasons for this deviation can be slightly different local conditions in the station surroundings, e. g., higher concentrations of biogenic volatile organic compounds such as monoterpene or isoprene. The ozone formation in the lower atmosphere is a highly complex interaction between VOCs and NO_x in the presence of sunlight [20]. Rural areas, especially such as surroundings of Rūgšteliškis, are usually NO_x limited due to the large quantity of trees that produce high levels of VOCs and, therefore, the concentration of ozone depends on the amount of NO_x in the atmosphere. Another reason can be different underlying surfaces in the surroundings of stations, which have substantial influence on the ozone destruction, especially at night and early in the morning. As mentioned above, the rate of ozone dry deposition to the water surface can be ten and more times lower, i. e., the ozone concentration near the sea should be higher than that inland under the same meteorological conditions.

The maximum values of the calculated parameters in the environment of both stations are presented in Table 1. The calculated values of E_0 and E_S at the Preila and Rūgšteliškis stations show some differences. It can be explained by different air turbulence intensity in the surroundings of the stations. Surface roughness effects on wind shear and turbulence profiles can be significant at heights up to 100 m [21]. Because

Table 1. Values of calculated parameters for the Preila and Rūgšteliškis stations: the maximum UV radiation intensity I_0, the air enrichment with ozone at nighttime E_0, the air enrichment with ozone at maximal turbulent mixing E_S, the daily air enrichment with ozone E_d, and the average ozone lifetime τ.

Station	Season	I_0, mW m^{-2}	E_0, μg m^{-3}h^{-1}	E_S, μg m^{-3}h^{-1}	E_d, μg m^{-3}day^{-1}	τ, h
Preila	Winter	1180	8.9	1.6	221	5.6
	Spring	4960	13.8	6.1	381	4.9
	Summer	6930	16.2	10.0	478	3.6
	Autumn	2930	8.7	5.5	243	5.2
Rūgšteliškis	Winter	1180	12.3	2.1	306	3.9
	Spring	4960	18.8	15.3	574	3.1
	Summer	6930	11.2	22.6	467	2.8
	Autumn	2930	8.5	11.0	276	3.7

Table 2. Correlation coefficients between I_0, E_0, E_S, Δh, and τ.

	I_0	E_0	E_S	τ	Δh	I_0	E_0	E_S	τ
	Preila					Rūgšteliškis			
I_0	1.000	0.949	0.966	−0.949	0.999	1.000	0.215	0.988	−0.989
E_0		1.000	0.847	−0.909	0.961		1.000	0.107	−0.318
E_S			1.000	−0.952	0.960			1.000	−0.955
τ				1.000	−0.959				1.000
Δh					1.000				

of low surface roughness on the relatively smooth water surface, the wind speed does not increase as much with height above the sea level as it does on land. The land surface roughness near the Preila station, where scrubs are predominant, is also much lower than near Rūgšteliškis where mature forest is widespread. For this reason, the air turbulence is more intensive and values E_0 and E_S are higher at the Rūgšteliškis station. All these factors determine the obtained ozone lifetime at two rural stations (Preila and Rūgšteliškis) having different values during the same season (Table 1). The estimated ozone lifetimes are consistent with the results found in other works [22–25]. The comparison of the estimated ozone lifetime under different rural conditions, e. g., with prevalence of different underlying surfaces, shows that ozone lifetimes differ from 1.3 up to 1.6 times. The ground-level ozone lifetime was estimated to be in the range of 3.6–5.6 hours at the rural Preila station and in the range of 2.8–3.9 hours at the Rūgšteliškis station during different seasons.

The linear regression method was applied to estimate correlation coefficients between I_0, E_0, E_S, Δh, and τ variables at both stations. Δh is the difference between the maximum and minimum mixing height at the Preila station. The obtained results are presented in Table 2.

As can be seen from Table 2, most of the correlation coefficients (indicated in bold) are statistically significant, at the significance level $\alpha = 0.05$. According to [26], if the number of paired observations is equal to 4 (see Table 1), the critical value of the correlation coefficient should be not lower than 0.950 at the significance level $\alpha = 0.05$. It means that the ozone enrichment maximum due to turbulent mixing is proportional to the maximum of the solar radiation intensity. That confirms that the solar radiation intensity can be used as the main parameter for the calculation of the ozone lifetime under rural conditions.

5. Conclusions

The new method for the estimation of the ground-level ozone lifetime under rural conditions during different seasons was developed. The method allows evaluating the ozone lifetime according to the discrepancy between the solar radiation intensity and duration, related turbulent air mixing height maximum, and the ozone concentration maximum during the day. The least-squares method was applied to determine the ozone decay rate and the air enrichment with ozone during the night hours and under maximum turbulent mixing conditions in the atmosphere. The ozone lifetimes were calculated using data from the Preila station. For the method validation, the same method was applied to the data from the other station Rūgšteliškis. The obtained ground-level ozone lifetimes were similar. They were in the range of 3.6–5.6 hours at the rural Preila station and in the range of 2.8–3.9 hours at the Rūgšteliškis station during different seasons.

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OZONO GYVAVIMO TRUKMĖS PAŽEMIO ORE MAŽAI UŽTERŠTOJE VIETOVĖJE NUSTATYMAS

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Santrauka

Ozono gyvavimo trukmė pažemio ore labiausiai priklauso nuo cheminių priemaišų koncentracijos ore, meteorologinių sąlygų bei vietovės paklotinio paviršiaus, ant kurio didžioji dalis jo suyra, fizinių ir cheminių savybių. Visų šių parametrų, nustatant ozono gyvavimo trukmę, įvertinimas yra labai sudėtingas ir keblus uždavinys. Straipsnyje pateikiamas metodas leidžia įvertinti ozono gyvavimo trukmę mažai užterštoje vietovėje, remiantis Saulės spinduliuotės intensyvumu ir trukme bei Saulės intensyvumo ir su juo susijusio turbulentinio oro maišymosi maksimumo ir ozono koncentracijos maksimumo per parą nesutapimu. Šis trukmės poslinkis atsiranda todėl, kad ozonas, šviečiant Saulei bei intensyvėjant turbulentiniam oro maišymuisi, kaupiasi ir jo koncentracijos ore maksimali vertė paros eigoje stebima vėliau nei maksimali Saulės spinduliuotė. Saulės spinduliuotės intensyvumas ir trukmė yra svarbiausi veiksniai praturtinant pažemio oro sluoksnį ozonu, nes, didėjant spinduliuotės intensyvumui, didėja ir turbulentinis oro maišymasis bei aktyvėja fotocheminės reakcijos. Saulės spinduliuotės intensyvumo kitimo matematinė išraiška gauta iš eksperimentinių Preilos stoties matavimo duomenų. Turbulentinis atmosferos maišymasis neišnyksta ir naktį, tik žymiai susilpnėja, todėl modelyje yra parametras, aprašantis pastovų pažemio oro sluoksnio praturtinimą ozonu visą parą, ir kitas parametras, atitinkantis ozono susidarymą esant Saulės spinduliuotei. Skaičiavimuose naudotas mažiausių kvadratų metodas. Panaudojus 2004–2005 m. eksperimentinius Preilos ir Rūgšteliškio stočių sezoninius ozono koncentracijos ore duomenis atlikta sukurto modelio patikra parodė gerą eksperimentinių rezultatų ir modelio atitikimą. Įvertintos ozono gyvavimo ore trukmės (3,6–5,6 valandos) Preilos foninėje stotyje yra ilgesnės nei Rūgšteliškio stotyje (2,8–3,9 valandos). Žinoma, kad ozono gyvavimo trukmė priklauso nuo azoto oksidų (NO_x), biogeninių lakiųjų organinių junginių (monoterpenas, izoprenas ir kt.) ir lakiųjų organinių junginių (LOJ) koncentracijos ore. Preilos stotis gali būti priskiriama vietovei, kur ozono koncentracijos dydžiui įtakos gali turėti LOJ kiekis atmosferoje. Rūgšteliškio stotyje eksperimentiniai matavimai rodo padidintą NO_x koncentraciją. Šios stotys yra vietovėse su labai skirtingu žemės paklotiniu paviršiumi, t. y. Preila yra ant Baltijos jūros kranto, o Rūgšteliškis – miškingoje vietovėje. Dėl didesnio paklotinio paviršiaus šiurkštumo Rūgšteliškio stoties aplinkoje vyksta didesnis nei Preilos vietovėje turbulentinis oro maišymasis ir didesnis pažemio oro praturtinimas ozonu. Šių vietovių išvardinti ypatumai ir lėmė skirtingus ozono gyvavimo ore trukmes.