Multi-model Assessment of Tropospheric Ozone Pollution Indices of Risk to Human Health and Crops, and Ozone Deposition in Ciuc Depression – Romania

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The Greenhouse Gas and Air Pollution Interactions and Synergies (GAINS) model quantified the relationship between the premature mortality and the long-term troposphere ozone concentration. Researches show that the elevated surface ozone concentration has an adverse impact on human health and vegetation also. In the present paper we examine the effect of ozone concentration with different indicators. Among the indicators on vegetation we examine the AOT40 and the W126. The limit values for these indicators according to the UNECE and the EU (2010) are 5000 ppb and 6000 ppb. The values of the indicators reported on Ciuc Basin ozone concentrations (AOT40, W126) does not cross the limits, but if we examine them in connection with the human health, it turns out that they exert an influence on the mortality. During the examination of the climate penalty factor a significant ozone increase can be reported in case of a possible temperature rise. This phenomenon is in accordance with the rate of mortality. The deterioration of the vegetation is caused by the ozone deposition, which may occur in dry or wet conditions. In this case we examine the wet deposition in relation with the coniferous woods of the Ciuc basin. Our results worked out in accordance with the ozone concentration indicators (AOT40, W126).

Keywords: tropospheric ozone, static stability, mortality, ozone deposition

The ground-level ozone (O_3) is one of the most important key pollutant gas, which has a major impact on human health and vegetation at regional and at global level [1-6].WHO (World Health Organization) (2003) reported that exposure to high ozone levels is linked to respiratory problems, such as asthma and inûammation of lung cells [3]. Several studies from previous years can prove that the air-polluting gases, including the ozone, also play a role in the development of various diseases (chronic bronchitis, emphysema, acute respiratory health effects, weaken the immune system) [7-9] and they are related to the mortality [8,10-12]. As it is deleterious to the human health, obviously it has an effect on the environment as well, a series of observations show that it can affect the agricultural yields, the viability of the vegetation and the health of animals [13,14]. This situation could dramatically worsen over the coming century [15].

The studied ground-level ozone is created by sunlight in the troposphere, involving the precursor is nitrogen oxides (NO_x), volatile organic compounds (VOC) and carbon monoxide (CO) [16]. The reaction of the CO with the hydroxide radical result carbon-dioxide and proton, which in reaction with oxygen and a third molecule produce hydrogen-dioxide. This in further reaction with nitrogen-monoxide create another nitrogen-dioxides in the atmosphere, accelerating in this way the formation of ozone [17]. This contamination, beside the natural ozone formation, rises mainly from anthropogenic activities and expanding biomass [18]. The further increase of ground-level ozone concentration, at regional or local level, will lead to further high exceedances [19]. The US EPA quantified these limits (United States Environmental Protection Agency) and the EU as 40 and 60 nmol/mol [20, 21]. In 2003, as consequence of the high temperature values, in 17 countries from Western and Central Europe were observed exceeded limits, leading to health problems and spectacular damage to the vegetation among others in France, Spain and Italy [22].

Researches show that since 1950 in the Northern Hemisphere the ozone concentration has been doubled [3,23]. According to the forecasts at the Northern Hemisphere the ozone concentration will increase with 5 nmol/mol value until 2030 and it could reach the 20 nmol/mol value until 2100 [15].

In the present paper we use several models based on the hour ozone concentration value. We discuss a wide range of currently used air quality standards for vegetation (AOT40 and W126) and human health ($SOMO_{35}$). We examine in addition the possible increase of the ozone concentration caused by the rise in temperature and its influence on deaths in the Ciuc basin, as well as the cause of the damage to the vegetation and the dry deposition of the ozone concentration to the pine forests.

Experimental part

Monitoring sites

The sites under survey are Miercurea Ciuc (662 m elevation), with 46°21’28.80"N latitude, 25°48,14’40"E longitude and Jigodin (717 m elevation), a suburban part of Miercurea Ciuc with 46°20’22.79"N latitude and 25°"48’26.95"E longitude; the distance between the two stations is 3.6 km. The geographical location of the Ciuc

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basin is similar to other bowl-shaped basins which produce significant temperature inversions [24]. The types of fog are characteristic to the locations under survey, especially during the winter [25].

Sampling

The NO_2, NO_x and O_3 data were under constant surveillance since 21 March 2012 until 20 March 2013, we used the month average data from 2007-2013 as well, which are registered by the regional automatic background monitoring station HR 01 from Jigodin and the Harghita County Environmental Protection Agency APM monitoring station destined to monitor the values of urban pollution.

Our measuring equipment's are the nitrogen oxide (ME9841B monitor Europe, US EPA, no. RFNA-1292-090) and the ozone analyzer (ME9810B Monitor Europe, Fotometric UV, US EPA, no. of reference EQOA-0193-091). The nitrogen dioxide analyzer measures continuously NO, NO_2 and NO_x values using chemi-luminescence method (in case of the measuring station from Miercurea Ciuc only NO_2 data were available in the period under survey), the ozone analyzer measures O_3 in the air using UV absorption method. We examine the air temperature using TS Thermometer sensor which can detect values between -30 and -50 °C. The equipment is installed two meter high from the ground. The level of the sun intensity is registered by the ORION – Mod SR-S sensor.

During this analysis, we have used validated data from Environmental Protection Agency of Harghita County, by this we managed to avoid inconsistent values. The data processing have been realized with hourly values for NO, NO_2, NO_x, O_3.

Results and discussions

Climate penalty factor and mortality

The high ambient temperature induces intensive photochemical reactions, which product is the tropospheric ozone. These phenomena can be described by the penalty factor which is related to the ration of oxygen/temperature.

It is defined as $\frac{\partial[O_3]}{\partial T}$ [26, 27].

For the evaluation of the penalty factor we used the temperature interval 19-37°C, and we selected groups with 3°C interval length. In this group we evaluated percentage values, which gives its proportion in that interval. This yields the corresponding scaling from lowest value ($\alpha = 0$) up to the highest value ($\alpha = 1$). $\alpha = 25\%$ means the quartile, $\alpha = 50\%$ corresponds to the median value.

The average of the slopes (5, 25, 50, 75, 95%) yields the penalty factor. The lowest steepness is at 95%, which means 1.47 μg/m³/°C, while the highest is at 5% corresponding to the value 2.51 μg/m³/°C. The average slope is at 2.05 μg/m³/°C, which is similar to the values which one finds in literature [26].

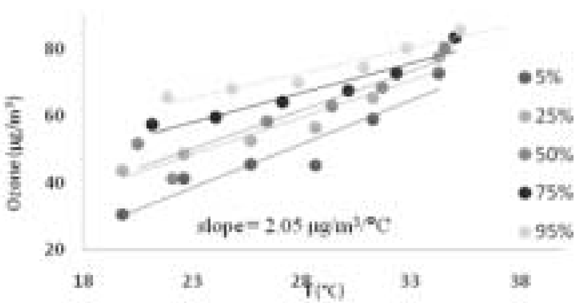

Fig. 1. The penalty factor based on the data of the Jigodin station

In Miercurea Ciuc there are lower values, which can be explained by the decrease of the industrial activity and by this the NO_x emission is also lower. In the same time emission due to the motor cars and the burn of biomass contribute to a partial rise of NO_x.

The global climate change expectedly leads to the rise of the over ground temperature and aggravate the air pollution [28]. The presented penalty factor also shows, that there is relatively high ozone concentration in Ciuc Depression.

The statistical analysis presented shows, that in 1°C increase in temperature leads to an increase of 2.05 μg/m³ O_3. This result indicates the importance of controlling of NO_x emission, which may lead to the decrease of photochemical smog and effects of climate exchange.

The mentioned over ground ozone is related to certain extent to the risk of unnatural death [29-31]. This can be described by a mortality relation related to ozone [12]:

$$\text{Mort} \sim \frac{1}{365} \cdot \text{Deaths} \cdot RR_{O3} \cdot O_3 \quad (1)$$

where: Mort – Cases of premature mortality per year Deaths – Baseline mortality (number of deaths per year);

O_3 – Population-weighted $SOMO_{35}$;

RR_{O3} – Relative risk for one percent increase in daily mortality per ppb 8-h maximum ozone concentration per day [12].

$$RR_{O3} = e^{\beta_{O3} \cdot \Delta O3} \quad (2)$$

where: β_{O3} - 0.0007 (derived from the WHO (2004) recommended value of 0.003 per 10μg/m³ O_3, converted to the risk rate for 1 ppbV of O_3);

ΔO_3 - $SOMO_{35}$/365, i.e. the average daily exceedance of the maximal 8 hourly average above 35 ppbV, averaged over 1 year, expressed in ppbV.

If we consider the proportionality constant to be one [12], than we get a value for number of persons 0.34 resulted by this evaluation, which corresponds to 0.0095% of unnatural life cease as partially due to ozone. We mention that this result is obtained assuming that other factors, which may act at the same time with ozone has neglect able effects on this rate.

Ozone air quality vegetation indices

The tropospheric ozone has a harmful effect on vegetation [1,9]. The measure of this effect can be presented with different indicators. In present work we tried to evaluate AOT40 and W126 values [3].

The AOT40 is defined as the sum of hourly daylight (>50 W/m² PAR) ozone volume mixing ratio (vmr) above 40nmol/mol [3,32,33]:

$$AOT40 = \sum_{i=1}^{n} [O_3 - 40]_i \quad \text{for } O_3 > 40 \text{ nmol/mol} \quad (3)$$

where O_3 is the hourly ozone vmr in nmol/mol and n is the number of hours with O_3 > 40 nmol/mol.

And the W126 is defined as the weighted sum of 24-hourly ozone vmr [3,32,34]:

$$W126 = \sum_{i=1}^{n} [O_3]_i \cdot w_i \quad (4)$$

where:

$$w_i = \frac{1}{1+4403 \cdot \exp(-0{,}126 \cdot O_{3i})} \quad (5)$$

and O_{3i} is the hourly ozone vmr in nmol/mol.

The prescribed limits has different values. The UNECE prescribe a 5000 nmol/mol (2004), the EU a value of 6000 nmol/mol (2010) in long term for limit value of AOT40 [35].

The periodic limit value for 3 month ($AOT40_{3m}$) corresponding to the months May-July is 9000 nmol/mol [34,35]. We have got a value of 931.3373 nmol/mol for the AOT40 value for the full year. Comparing this value to the limiting one, one can say that it is considerably below of limiting value. During an intensified vegetative production (May-July) the value of $AOT40_{3m}$ will decrease to 84.5383 nmol/mol. For the productive period of the forest (April-September) $AOT40_F$ value rises to 222.3448 nmol/mol. The values of W126 shows a similar tendency, for the full year it results in 2090.3478 nmol/mol, in May-July period 503.7117 nmol/mol, and for the period April- September 974.7843 nmol/mol. There is no explicit described limit value for W126 [36,37].

The dry deposition of ozone close to the ground

The emitted substances in the atmosphere after a certain time may be transformed and a part of them can become removed from it, by dry or wet deposition [38-45]. In our work we studied the dry deposition of the ozone close to ground for conifers in the Ciuc Depression. The harmful effect of ozone have been already studied for pine trees [46,47], the absorbed ozone on gas exchange openings oxidize the chloroplasts in an inadequate way [48], by this decreasing the productivity and sub serving the ageing [49-52]. The measure of damage is given by the exposure to the concentration, and in a reduced extent the flux of ozone [53].

The deposited amount is related to the different indicators (AOT40, SOM0, W126) [52,54,55], from which the AOT40 and W126 have been already evaluated. The AOT40 a dose given about the sum of total ozone flux [56].

To describe the deposition process, we need the deposition mass flow density (deposition flux) F_t, and the deposition speed, v_d [42-45,54-59].

$$F_t = v_d \cdot c_r \tag{6}$$

where F_t is the ozone flux, v_d is the deposition velocity, and c_r is the concentration of ozone.

$$v_d = \frac{1}{R_a + R_b + R_c} \tag{7}$$

where: R_a, R_b and R_c are the aerodynamic resistance, the quasi-laminar boundary layer resistance, and the canopy resistance, respectively.

For determination of aerodynamic resistance we should find the value L of Monin-Obukhov lengths, and the value of friction velocity [60-62]

$$u_* = k \cdot u \left[\ln\left(\frac{z-d}{z_0}\right) - \psi_m\left(\frac{z-d}{L}\right) + \psi_m\left(\frac{z_0}{L}\right)\right]^{-1} \tag{8}$$

$$L = -u_*^3 \frac{\rho_a \cdot Cp_a \cdot T}{k \cdot g \cdot q_H}, \tag{9}$$

where $\psi_m(\xi)$ is the integral from of universal stability correction functions for the momentum, ρ_a is the air density, Cp_a is the specific heat at constant pressure, T is the air temperature K, k=0.4 is the von Karman constant, g=9.80665 m s^{-2} is the acceleration of gravity, q_H is the sensible heat flux, z, z_0 and d are the reference height, the roughness lenght, and the displacement height, respectively.

The empirical relations related to boundary layer resistance one can find in literature [39]. Further terms in deposition velocity are determined by R_{st}, R_s and R_{cut} (stomatal, surface and the cuticular resistances, respectively) [45].

$$R_c = \frac{1}{\frac{1}{R_{st}} + \frac{1}{R_s} + \frac{1}{R_{cut}}} \tag{10}$$

The stomatal resistance empirical formula [61]:

$$R_{st} = \frac{1}{G_{st}(PAR) f_t(t) f_e(e) f_\theta(\theta) f_{D,i}} \tag{11}$$

where $G_{st}(PAR)$ is the unstressed canopy stomatal conductance, a fanction of PAR (photosynthetically active radiation):

$$G_{st}(PAR) = \frac{LAI_s}{r_{st}(PAR_s)} + \frac{LAI_{sh}}{r_{st}(PAR_{sh})} \tag{12}$$

and the $r_{st}(PAR_s)$, $r_{st}(PAR_{sh})$ is calculated whit the following form:

$$r_{st}(PAR_s) = r_{st,min}\left(1 + \frac{b_{st}}{PAR_s}\right) \tag{13}$$

$$r_{st}(PAR_{sh}) = r_{st,min}\left(1 + \frac{b_{st}}{PAR_{sh}}\right) \tag{14}$$

The LAI_s, LAI_{sh} are the total sunlit and shaded leaf area indices respectively, and PAR_s, PAR_{sh} are photosynthetically active radiation received by sunlit and shaded leaves, respectively [62]. The $r_{st,\,min}$ is a reciprocal value of the so-called maximum stomatal conductance, and b_{st} is a plant species dependent constant. The values $r_{st,\,min}$ = 150 and b_{st} = 44 [41,54].

The $f_t(t)$, $f_e(e)$ and $f_\theta(\theta)$ describe the effect of temperature (eq. 10), vapour pressure deficit (eq. 12) and soil water stress on stomata (eq. 13), respectively, and $f_{D,i}$ modifies the stomatal resistance for ozone 0.625.

The temperature stress function is described by the following equation:

$$f_t = \frac{T - T_{min}}{T_{opt} - T_{min}} \left(\frac{T_{max} - T}{T_{max} - T_{opt}}\right)^{b_t} \tag{15}$$

where:

$$b_t = \frac{T_{max} - T_{opt}}{T_{max} - T_{min}} \tag{16}$$

T_{min}, T_{max} and T_{opt} are the vegetation dependent minimum, maximum and the optimal temperature, respectively. These term values are T_{min} = -5°C, T_{max} = 40°C and T_{opt} = 15°C [45].

The stress of the vapor pressure deficit can be parameterized by the following form:

$$f_e = 1 - b_e(e_s - e) \tag{17}$$

Here b_e is a vegetation dependent constant, e and e_s are the water vapour pressure and the saturated water vapour pressure, respectively.

The water stress function is parameterized using soil water content (θ):

$$f_\theta = \begin{cases} 1, \text{ha } \theta > \theta_f \\ \max\left\{\frac{\theta - \theta_w}{\theta_w - \theta_f}, 0{,}05\right\} \text{ha } \theta_w < \theta \le \theta_f \\ 0{,}05, \text{ha } \theta \le \theta_w \end{cases} \tag{18}$$

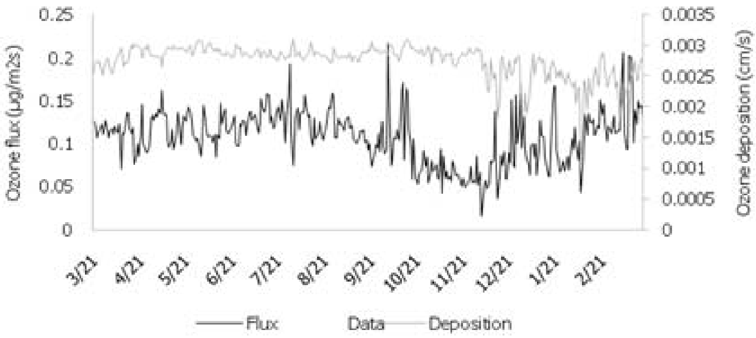

Fig. 2. The daily ozone flux and ozone deposition in the period under consideration

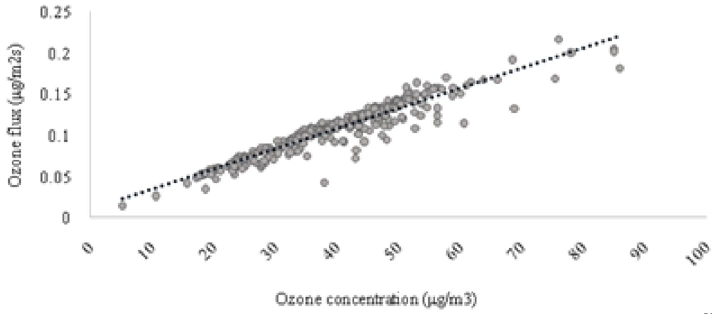

Fig. 3. The relation between the ozone concentration and ozone flux

where θ_w and θ_f are the wilting point and the field capacity soil moisture contents, respectively.

The cuticular resistance, R_{cut}, is calculated by an empirical [63]:

$$R_{cut} = \frac{R_{cut,0}}{e^{0,03\varphi} LAI^{0,25} u_*} \quad (19)$$

where φ is relative humidity (in percentage), $R_{cut,0}$ = 2000, and LAI = 6.2 [41,63,64].

The surface resistance, R_s, for ozone deposition is 300 s m^{-1} [41].

In the period March the 21th 2012 and March 20th 2013 in a similar way to other studies, the high fluxes of ozone appears at the active period of vegetation of the year, in our case from May to October [65]. During the winter and the spring we also can see high flux values, although most of them shows lower tendencies relative to the other periods, mentioned above [66] (fig. 2.).

The positive values of the flux shows, that there is a reception by vegetation [66], which are in our case in the interval 0.015 cm s^{-1} and 0.03 cm s^{-1}, and these values are relatively small and do not produces visible damages – supported also by the AOT40 and W126 indicators [56]. In the same time the deposition my cause growth problems.

During the year the deposition, in a similar way to the tendency of the flux, shows lower values in winter and spring, while in other periods can be considered to be more or less uniform [53] (fig. 2). In this process, the activity of the vegetation has a role, and the bad absorption of the icy relief [67].

The regulation of ozone concentration which is in taken by gas exchange openings is realized by the ozone flux [66]. Between the ozone concentration and ozone flux here is a 0.944 Spearman rang correlation which is illustrated in figure 3. [67].

Between the deposition and the flux there is a linear relation [53,65], although in some cases one can find low flux with high deposition (fig. 4.) and high ozone concentration (fig. 3.). Usually this is due to the high ozone concentration and high temperature. At high temperature with high vapour pressure deficit, the gas exchange openings become closed, and the deposition decreases [53,66-69].

In our case the low values of fluxes occur at 01:00 – 08:00 o'clock, while the high values are at 09:00 – 20:00 [53].

In these periods the values of deposition are different, at 01:00 – 08:00 is high the deposition and in the interval 09:00 – 20:00 is low. These values are opposite with the values of ozone concentration, which are high in the afternoon and are low in the morning [67]. Taking into apart the hourly values to seasonal ones, one can see that the flux has a similar behavior in all periods, which means in summer maximal and in autumn minimal, rise in the spring and decay during winter [65] (fig.5.). In the summer when the activity of vegetation is intensive, the intake of ozone in gas exchange opening becomes important, by this the value of ozone deposition becomes considerable [39,57,65,70 -73].

Based on the present studies, in the winter the weak ozone deposition is due to the frozen and snowy layers, which are bad absorbents [68].

During spring and summer the maximum values of the deposition occur in morning hours (06:00-09:00), in this interval the gas exchange openings are the most active, later for limitation of the water loss, their activity decreases. The hourly variations follows the season is changes. In the afternoon hours in autumn and in summer the values are

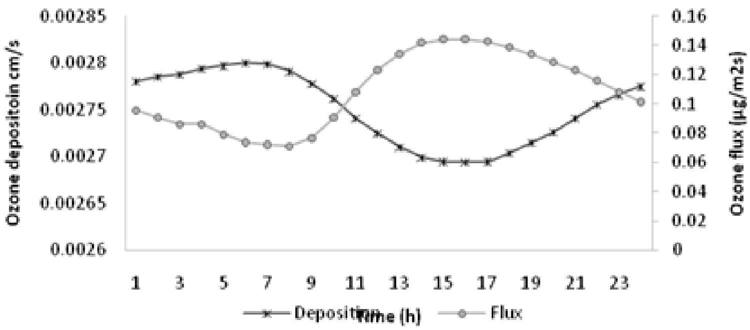

Fig. 4. The ozone deposition and the ozone flux hourly average during the day.

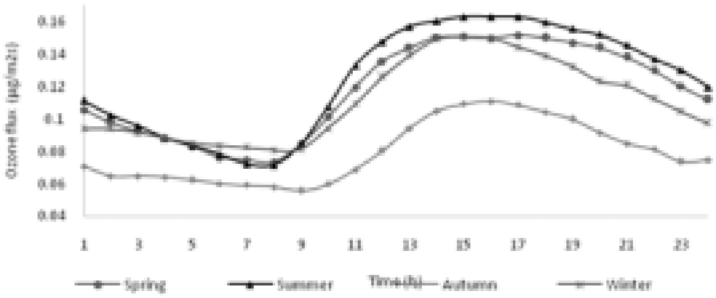

Fig. 5. The ozone flux in each season

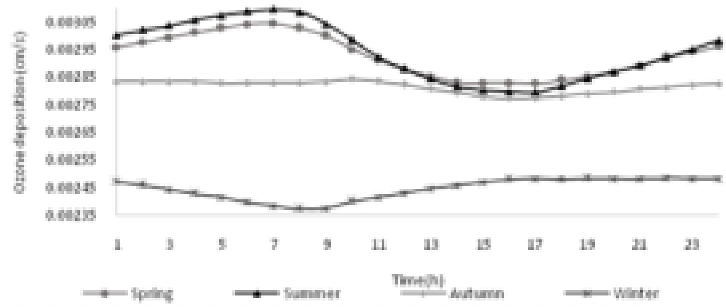

Fig. 6. Ozone deposition in each season

low, the ozone deposition shows increase in spring. In evening hours in three seasons there is an increase of ozone deposition [67]. Wintertime the deposition is almost constant with a slight uniform increase at evening hours [53]. The ozone flux contrary to the ozone deposition in the morning shows fast increase (08:00-10:00), while in the evening it progressively decreases.

Conclusions

Because of the relatively closed, bowl form of Ciuc depression one can observe the presence of microclimate. This microclimate is slightly influenced the external air fronts, which is practically neglectable, by this the static stability – in terms of Brunt-Väisäla frequency – has an enhanced impact on the polluting substances. In our study we played attention on the deposition of the ozone close to ground.

Based on the research in this field, the high concentration of ozone close to ground has harmful effects on human health, and it may influence the unnatural death. If the ozone concentration exceeds certain values, than the effect on life cease yields a finite value slightly above zero. This has a very low influence on death data, although a 1°C rise on temperature, may bring to the increase of the mortality indicator. Other indicator is the penalty factor, its investigation showed, that 1°C increase in temperature, leads to 2.05$\mu g/m^3$ O_3 increase in the atmosphere of Miercurea Ciuc.

This result shows the necessity of the control on NO_x emission, which is related to the ozone concentration. By the increase of ozone concentration we get a more polluted atmosphere which also has an effect on vegetation. The values of AOT40 and W126 shows that the concentration is not so high to harm the vegetation, although the deposition values shows that with the increase of concentration the harmful effect on vegetation may become enhanced.

The most enhanced ozone deposition have been occurred in morning hours when the gas exchange openings are active, while the lowest values of deposition was in afternoon, when the gas exchange openings have a minimal activity.

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