EVALUATION OF CUMULATIVE AIR POLLUTION IN RIGA AND LIEPAJA WITH CUMULATIVE POLLUTION INDEX METHOD

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Abstract
Cumulative air pollution – synergy between different pollutants and environmental factors is one of the hardest to evaluate factors in the air quality monitoring field. The evaluation of cumulative effects is hindered by a lack of verified analytical frameworks. Currently used methods are relatively simple and use statistical models with small fixed number of pollutants in association with different factors. There is almost no one solution for direct, on-site cumulative effect measurements. The alternative is the use of Cumulative Pollution Index (CPI) method – solution designed for cumulative effect calculation from bioindication and air quality measurement data. However, this method is completely new and has never been used in the air pollution evaluation activities. Therefore, the objective of this study was to evaluate the opportunity to use CPI method as a cumulative pollution evaluation tool in air quality monitoring by doing measurements of cumulative effect in several air quality measurement stations in two cities of Latvia – Liepaja and Riga. Results show that Cumulative Pollution Index method is not only usable in air quality monitoring as a tool for cumulative effect evaluation, but can reveal new facts about air pollution and ways how it affects human and ecosystem health, – such climatic and environmental factors as humidity and temperature are more important than interactions between individual pollutants and can be considered main elements in forming of cumulative pollution impact.
Key words: air pollution, cumulative effect, bioindication, Cumulative Pollution Index.

Introduction

Air pollution is one of the most actual environmental problems in the world. Increasing traffic density and energy consumption lead to increased pollution causing substances emissions in ambient air. It is a significant risk factor for multiple health conditions, including lung cancer, respiratory and heart diseases. Therefore, it is important to keep up with the latest data about the actual air quality to react timely and initiate appropriate environment management procedures when the pollution levels rise too high (Snyder et al., 2013).

For these purposes, air quality monitoring is carried out. It is a regular and continuous collection of information about air quality to prevent hazards associated with pollution. Usually, as a monitoring technical solution, automated measurement stations are used. They measure concentrations of various substances in nonstop mode – ranging from gaseous pollutants, like nitrogen oxides (NOx) and ozone, to particulate matter. Results are compared according to normative standards to determine if the actual pollution level is a threat or not. However, one thing is to control individual substances – another is the actual impact, caused by synergy of different pollutants and environmental factors. Latest research shows that there exists a hidden threat called cumulative effect – synergy between different pollutants and environmental factors which produce greater impact on living organisms than the same substances in separate action. For example, ozone mixing with other pollutants leads to increased effect on human health (Mauderly and Samet, 2009). Such cumulative effects are very complex and depend on many factors – weather, seasonality, exposure duration, etc. (Stylianou and Nicolich, 2009; Su et al., 2012). Therefore, it is hard to evaluate them.

As cumulative risk assessment of real-world mixtures is hindered by a lack of verified analytical frameworks (Callahan and Sexton, 2007), there are only a few methods for cumulative pollution evaluation. They are relatively simple and use statistical models with a small fixed number of pollutants in association with different factors. An example of such methods is Su et al. (2009) cumulative environmental hazard inequality index (CEHII) which assesses exposure to multiple air pollutants within different racial-ethnic groups and socioeconomic positions in Los Angeles. Another approach is the definition of cumulative pollution as a difference of living organisms' health and measurement results in the same pollution level as it is done in Cumulative Pollution Index (CPI) method – a solution designed for cumulative effect calculation from bioindication and air quality measurement data (Kalniņš, 2012).

Bioindication is a pollution evaluation method which uses living organisms as indicators of pollution level and environmental quality. By applying methods of bioindication, it is not possible to make measurements of air pollutant concentrations as with sensors, though it is an effective tool to evaluate exposure, dose and bioaccumulation – factors which are directly related to cumulative effects. On the contrary, air quality measurements is the main source of information about pollution causing substances in the air – they can't determine measured pollutants' effect on living organisms, but they can detect individual pollutants and their amount in ambient air (Snyder et al., 2013).

By merging these two approaches – measurements and bioindication, on-site measurements of cumulative effects can be done. However, CPI method is completely new and has never been used in the air pollution evaluation activities (Kalniņš, 2012).

Therefore, the objective of this study was to evaluate the opportunity to use CPI method as a cumulative pollution evaluation tool in air quality monitoring by doing parallel measurements of cumulative effect in several air quality measurement stations in Latvia.

Materials and Methods

The study was carried out in three Latvian national air quality monitoring network sites, where automated measurement stations are placed:

In forest, near the ruins of South fortification of Liepaja (56º 28'48,41''N; 21º 00'01,06''E), in weight and size as similar as possible, lichen samples were collected and placed in perforated plastic containers – then delivered to the chosen air quality measurement sites and placed on automated monitoring stations in height of measurement equipment. To protect lichens from external factors during the transportation, perforated containers were placed into another – airtight containers. Sampling site was chosen according to the pollution dispersion modelling done by the municipality of Liepaja, which shows that on this site the air quality can be described as clean city air (Estonian, Latvian & Lithuanian Environment, 2004).

Three lichen species were used: foliose lichens *Xanthoria parietina* and *Parmelia sulcata* as well as fruticose lichen *Ramalina fraxinea*. They were chosen from different sensitivity groups to exclude specific responses to individual pollutants and environmental factors:

- *Ramalina fraxinea* – sensitive to almost all air pollutants (Nimis et al., 2002);
- *Parmelia sulcata* – intermediate SO_2 tolerant (Peterson et al., 1992; Hawksworth and Rose, 1970) while sensitive to other pollutants, for example, O_3 (Peterson et al., 1992; Ross and Nash, 1983);
- *Xanthoria parietina* – pollution tolerant species which is absent only in high pollution levels (Hawksworth and Rose, 1970; Perkins and Millar, 1987b).

The duration of the study was 12 months – from 01.02.2013. to 01.01.2014.

Chemical analysis of lichen samples

Each month, the containers with samples were removed from the monitoring stations, placed in airtight containers again and delivered to the Laboratory of Plant Biochemistry, Institute of Soil and Plant Science, Latvia University of Agriculture.

As it is possible to determine the pollution impact on lichens by chlorophyll and pheophytin ratio (Riddell et al., 2012; Tretiach et al., 2007; Hauck et al., 2003), and this approach is used in the CPI method, in the laboratory these biochemical values were measured with spectrophotometer Perkin Elmer Lambda 25.

For extraction of both necessary pigments, lichen samples were weighted, placed in 5 ml Dimethylsulphoxide (DMSO) and heated at a temperature of 65 °C for 45 minutes. Then the obtained solution was cooled, inserted in a spectrophotometer, and measured chlorophyll and pheophytin optical densities – 415 and 435 nm wavelengths, according to Ronen and Galun method (1984). To ensure that heavier and greater lichens with more pigment content do not influence the results, they are expressed in weight per optical density of solution (g/OD).

Table 1

Automated air quality measurement stations, used in cumulative pollution evaluation

Monitoring site	Coordinates	Measurement technology	Measured pollutants
Riga, Brivibas street 73	56°57'32", 24°07'34,03"	DOAS*; OPSIS/SM200 „ADAM"	SO_2, NO_2, O_3, benzene, toluene, PM10, PM2.5, Pb, Cd, Ni, As, Benzo(a)pyrene, PAO
Riga, Valdemara street 18	56°57'27,0", 24°06'57,05"	HORIBA traffic pollution measurement station	NO_2, NOx, NO, O_3, CO, PM10, benzene, toluene
Liepaja, Kalpaka street 34	56°31'31'', 21°00'13''	DOAS*; OPSIS/SM200 „ADAM"; HORIBA; diffusion tube	SO_2, NO_2, NO, O_3, CO, benzene, toluene, PM10, PM2.5, Pb, Cd, Ni, As

* Differential Optical Absorption Spectroscopy

CPI index calculation

Using the CPI method, cumulative effect is calculated as index from bioindicator samples health condition and air pollution measurement data using CPI equation (Kalniņš, 2012) with latest additions (2014) which makes it compatible with chlorophyll and pheophytin ratio approach:

$$CPI = \frac{\left\{\left[\left(\sum_1^{ns} P\right)/\left(\sum_1^{ns} C\right)\right]\times 100\right\}}{\left\{\left[\left(\sum_1^{np} C_p\right)/\left(\sum_1^{np} BP_p\right)\right]\times 100\right\}} \quad (1)$$

where:
CPI – cumulative pollution index;
Cp – concentration of pollutant p;
BPp – breakpoint of pollutant p concentration (according to normative);
np – number of pollutants;
ns – number of lichen samples;
C – total amount of chlorophyll and pheophytin in sample;
P– pheophytin amount in sample.

The obtained chlorophyll and pheophytin values are placed in equation 1. (sum of g/OD 435 and 415 nm as C; g/OD 415 nm as P) and together with the air quality measurement data calculated CPI index value.

It is relative, unitless value – the further from 1 as the point of equality between pollution and according to health condition, the greater cumulative impact. Since lichens are living organisms, there are possibilities of natural, pollution not-related pigment changes in them, and therefore, according to the instructions of CPI method usage, it is advisable to determine the threshold value when exceeding it the result is considered as detection of cumulative effect. In this study, as the threshold was chosen value 1 – base threshold, as it is described in CPI mathematical model (Kalniņš, 2012).

Results and Discussion

In this study, from all measured pollutants, 4 were used – SO_2, NO_2, O_3, CO, because they have clearly defined breakpoint values as they are specified in the main air quality normative act in Latvia – Cabinet Regulation No. 1290 „Regulations Regarding Ambient Air Quality" (as of 03.11.2009). Other pollutants, such as benzene and toluene have only breakpoint values related to calendar year, therefore their compliance with the air quality standards can't be evaluated in short term study like this.

According to measurement specifics and data accessibility, SO_2, NO_2, O_3 were used in cumulative impact evaluation process in Liepaja and Brivibas street, and SO_2, NO_2, O_3, CO in Valdemara street. Results – the obtained CPI values – are shown in Table 2.

In Liepaja the threshold is exceeded regularly with peak value in July. A bit different, but similar situation is in Brivibas street, Riga – threshold is exceeded in February and summer months, starting from June and ending in September. The peak is also in September and later CPI values gradually slip below the threshold (Fig 1.).

Results from Valdemara street are completely different – February also is above the threshold, but

Table 2

Monthly measured Cumulative Pollution Index (CPI) values

Month	Measurement place		
	Liepaja	Riga, Valdemara str.	Riga, Brivibas str.
February	1.273	1.140	1.077
March	1.008	0.844	1.009
April	1.106	0.879	0.966
May	1.104	0.877	0.968
June	1.094	0.742	1.308
July	1.393	0.940	1.309
August	1.222	0.928	1.086
September	1.105	0.915	1.507
October	1.142	0.801	1.061
November	1.194	1.449	1.028
December	1.159	1.361	0.976
January	1.054	1.077	0.965

CPI values are unitless – greater number means greater cumulative impact further cumulative effect is not detected; then, in November there is a peak and the cumulative effect decreases towards January. It is interesting that in some months – June and October, – CPI value is significantly lower than the threshold. As it is unlikely that in some circumstances the air pollution can become more health-friendly, this can rather be associated with natural changes in the amount of pigments in lichens. Therefore, it confirms the need for threshold approach in using CPI method.

In both cities Liepaja and Riga, the cumulative effect maximum is observed in summer months, except Valdemara street, where the CPI peak value is in November (Fig 1.). Therefore it is possible to propose a hypothesis that two of the main cumulative effect building factors are humidity and temperature, because in summer rainfall usually is higher and the air temperature also is significantly higher than the rest of the year. The fact that the cumulative effect in Liepaja is above the threshold almost all year, indirectly confirms this assumption, because due to closeness to large water masses - the Baltic Sea, Trade Channel and the Lake of Liepaja, the daily average relative humidity in the city is one of the highest in Latvia – 82%.

To determine the exact cause of observed cumulative impact variations, a more detailed and larger scale research is needed.

To better understand the obtained results and cumulative pollution forming factors, the CPI values can be viewed by their components separately – comparing bioindication measurement results with the overall pollution level changes (Fig 2.). Figure consists of two kinds of values – ‘pollution level’ which is

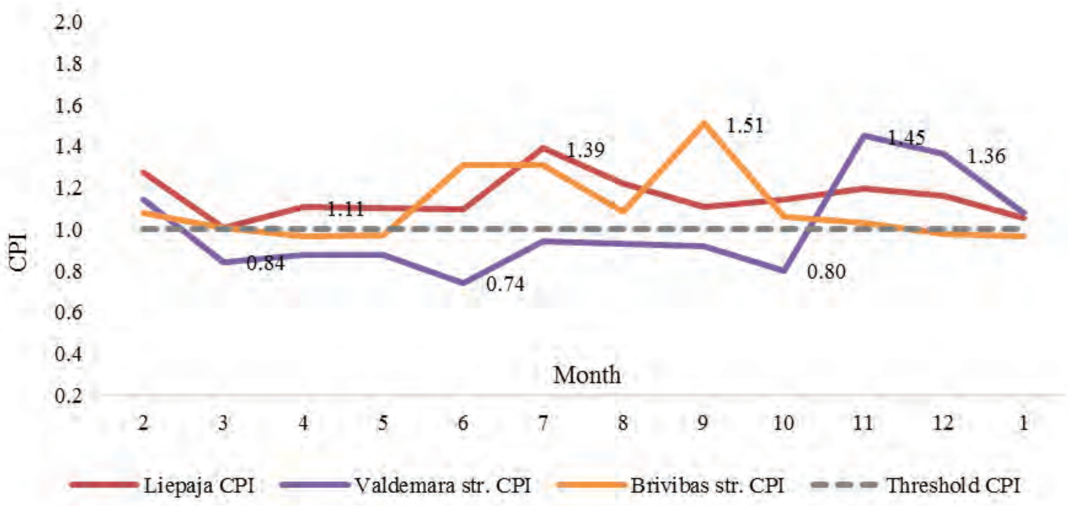

Figure 1. Monthly Cumulative Pollution Index values comparison with threshold (with numbers, only extreme values are shown).

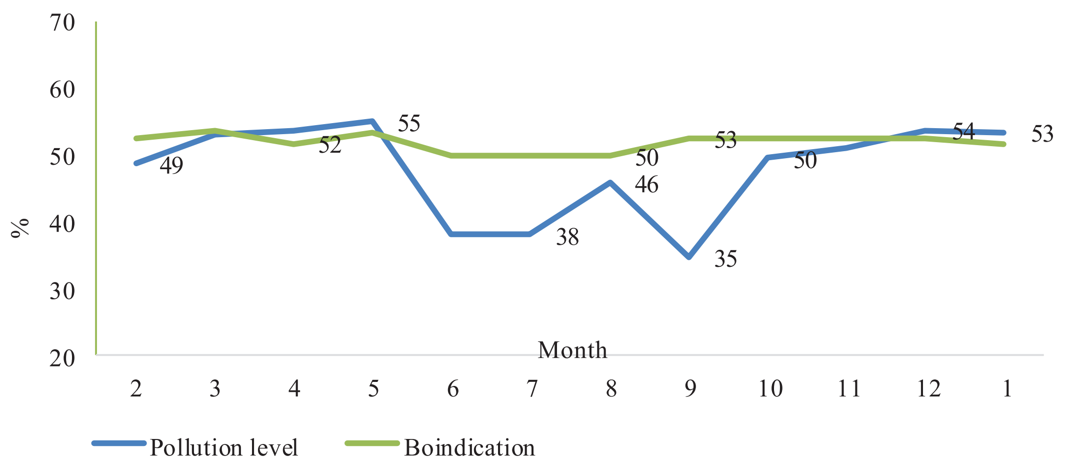

Figure 2. Variation of Cumulative Pollution Index forming components. Results from Brivibas str. in Riga (with numbers, only extreme values are shown).

measured values percentage from breakpoints, and 'bioindication' which is chlorophyll percentage from the total chlorophyll and pheophytin amount.

Such a comparison shows that the air quality measurement data and living organisms' health in the same pollution level not always are the same – often they vary independently from each other. For example, in results from Brivibas street, there are two points when the pollution measurement results are quite opposite to the bioindication data – in August the overall pollution level rises, while its impact on the indicator organisms do not change (pollution level from 38 to 46%; bioindicators damage the same 50%). In September, on the contrary, the pollution level decreases while its impact slightly increases (pollution level 35%; bioindicators damage 53%).

The overall trend in this example (Fig 2.) is that the pollution impact is relatively steady, while pollution causing substances concentrations in ambient air vary in relatively large range. It again raises assumption that in the cumulative pollution evaluation the climate and environmental factors play a more important part than previously known, and the interactions between individual pollutants is only a small part in the cumulative impact structure.

Therefore, it can be concluded that the Cumulative Pollution Index method is not only usable in air quality monitoring as a tool for cumulative effect evaluation, but can reveal new, previously unknown facts about air pollution and ways how it affects human and ecosystem health.

Conclusions

1. During the study, the cumulative effect in Liepaja air quality monitoring site is detected all year long, except in April, while in Riga only in summer (Brivibas str., June to October) and some autumn months (Valdemara str., November to December).
2. In both cities – Liepaja and Riga, the cumulative effect maximum (CPI value 1.3 – 1.39) is observed in summer months, therefore it is possible to propose a hypothesis that two of the main cumulative effect forming factors are humidity and temperature.
3. The study demonstrates the importance of climatic and environmental factors over interactions between individual pollutants as the main elements in building of cumulative air pollution impact – in some months living organisms' health worsens more than increases the overall pollution level (pollution level 35%; bioindicators damage 53% – in September in Brivibas str., Riga).
4. To determine the exact causes of cumulative impact variations, a more detailed and larger scale research is needed.
5. Cumulative Pollution Index method is usable in air pollution monitoring, because it can detect not only the cumulative impact, but can also reveal new facts about the air quality, thus improving understanding about pollution and its impact on living organisms.

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