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VALIDATION OF INDIRECT ASSESSMENT OF ANTHROPOGENIC IMMISSION LOAD

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Abstract. The assessment of anthropogenic immission load of area is important indicator in the process of immission participation determination to agricultural crop detraction. Despite the rapid concentration detraction of all immissions in the air environment dominates the opinion, that it continues in latent scale of damaging in the economically significant scale. Demonstration of this effect scale is economically and technically difficult. That is why the indirect methods of load assessment are developed, whereas the level of their validity is the cardinal inefficiency. Determination of concentrations of pollutants recognized methods are costly and labour-consuming and are also linked to energy networks. In the present work we follow the content of SO_2 in the atmosphere because of their greatest contribution to atmospheric pollution. Their objectives is based on selected methods simply set the load area of sulphur and nitrogen transformation of the volume concentration of SO_2 the deposition rate and replace the costly and labour intensive methods. In this paper we compare two direct methods sorption-cumulative and chemiluminescence. The amounts of SO_2 observed sorption-cumulative method in the individual periods was several times higher than the amount measured by AMS Leles. Results showed relatively large differences between different methods. The above methods can replace costly and labour intensive methods.

Key words: air pollution, imissions, methods for measuring sulfur dioxide

INTRODUCTION

The problem of immission influence on the agricultural crops (forest vegetation and generally on ecosystems) by reduction of air pollution concentrations is not so important. The situation in the last 50 years is gradually improving, though the influence of old environmental load still occurs and it is necessary to monitor and evaluate them. New methods and technologies development cause reduction of the state of emission exhala-

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tion into the atmosphere and consecutive reduction of immission influence on the Earth surface. In present time in Slovakia does not exist any special basis for quantification of damages caused by immissions considering ongoing immission situation. It is used in different modifications the methodology of ÚVTIZ č. 12/1992 [Neměc et al. 1992] and actually there are the doubts about negative effect of immissions on ecosystems, respectively presented assignments are not relevant. The research for agricultural purposes practically does not exist and there are known only the results of forest research only in other basis as it was used in the past.

Sulphur dioxide SO_2 in the past was the most serious air pollutant and identifier of pollution, considering the evolution in the last decade lost this position. It is the consequence of measurements agreed in the frame of the international agreements (Agreement of ECE UNO about remote air pollution transmigrant through the state's boundaries – Genova 1979, Protocol to Agreement about reduction of sulphur emissions at least about 30% – Helsinki 1985, Protocol about next reduction of sulphur emissions – Oslo 1994 – reduction about 60% up to the year 2000, about 65% up the year 2005 and about 72% up to the year 2010 in comparison with the year 1980, Protocol about reduction of acidification, eutrophication and ground ozone – Goteborg 1999 – emissions reduction SO_2 up to the year 2010 about 80% in comparison with the year 1990).

Slovak Republic after subscription of Genova Agreement of ECE UNO about remote air pollution obligated to fulfil the protocol about long term financing of Cooperating programme for monitoring and evaluation of remote transmission of pollution in the Europe EMEP (environmental Monitoring and Evaluation Programme). Systematic monitoring is required for local air pollution, for pollution of ground atmospheric layer of the cities and industrial areas. The main pollution sources are local energetics, industry, automobile transport and heating. They influence especially in primary form. Their concentrations show significant daily and season changes caused by local, topographic and meteorological factors. Industrial technologies are the air pollution source especially metallurgy, chemistry and production of building materials. It fills up, that unsuitable consequences of air pollution in the most part of Slovakia multiplied its segmentation.

Qualitative break reacting on improving of immission situation for harmful SO_2 was the suggestion of critical level of SO_2–S as a yearly average (Draft Manual for Mapping Critical Levels/Loads, UN ECE, 1990) with evaluation for ecosystems: forest 10,0 natural vegetation 10,0 agricultural crops 15 $\mu g \cdot m^{-3}$, when: critical level is the highest tolerated concentration of injuring, which already does not cause ecosystem abuse. Presented level was numerically identified with public notice of the Ministry of Environment from 29th Nov. 2002 about air quality number 705/2002 of the Statue effectives up to 31st May 2010, when the limit value of average concentration of SO_2 is 20,0 $\mu g \cdot m^{-3}$. In actual public notice from 12th August 2010 about air quality number 360/2010 of the Statue – Annex 11 presents the same limit values and class limits tolerance for sulphur dioxide (SO_2) as it is in the last public notice 705/2002 of the Statue [Kalúz et al. 2010].

Modelling in the GIS environment is advanced and effective tool for the creation and usage of the models with the goal of the research, predict and know the spatial chronological and functional aspects of geographical sphere [Varga et al. 2013].

MATERIAL AND METHODS

Characteristics of monitoring area

The locality of Vojany power station was determined for monitoring of air sulphur concentration. In presented immission locality is built automatic measuring station (AMS) Leles (operated by the ENVITECH company, sponsored by SHMU Bratislava, the results verified by propriate OÚ ŽP) used for measurement of the total pollutant concentrations. In presented area was placed meteorological rack (Fig.1). These two measurement equipment evaluate the data of SO_2 amounts measured by two direct methods – sorptive--cummulative and ultraviolet-fluorescence methods.

SORPTIVE-CUMMULATIVE METHOD

It was chosen sorptive-cummulative method for determination of area load by the sulphur. This method belongs among direct methods of measurement according to ČSN 038211 (method elaborated by SVÚDM-Praha, Běchovice) determined for long-term measurement of SO_2 concentrations and its compounds in the areas, where is monitored the level of atmospheric agresivity. The result value of sorptive amount of SO_2 (including sorpted elements of SO_3, H_2SO_4 and soluble sulphates) for areal unit of strong alkalized surface of filters presents immediate aggressive atmospheric influence.

Filtration laminaes absorbing sulphur allied substances are placed on the rack prepared according the parameters presented in ČSN 03 8110.

Preparation of filtration laminaes and determination of SO_2 by measuring analyse was realised at the Department of Chemistry of Slovak Agricultural University in Nitra.

Fig. 1. Meteorological booth with filtering plates

The method of direct measurements by isotherm air-conditioned container LU 3000

The analyser Thermo Electron Corporation, model 43 c works on the principle of ultraviolet fluorescent method based on emissions of radiation by SO_2 molecules excited by UV radiation for their passing into basic energetic state. The intensity if fluorescent radiation is proportional to the number of SO_2 molecules in detection volume and concentration of SO_2.

The analyser provides the variation compensation of output apparatus signal caused by oscillation of internal apparatus temperature as well as the compensation on pressure changes in fluorescent tank. SO_2 concentration is directly measured in the units of the volume portion, if the analyser is calibrated according to the standard with concentration expressed as volume portion. The final results are expressed in $\mu g \cdot m^{-3}$ after recalculation in evaluation system of standard recalculated coefficients.

The methods of indirect measurement

Mathematic model of dispersion of pollution materials in the air Win Modim version 4.11 elaborated by the ENVItech s.r.o. company Trenčín on the base of methodology ISC2 EPA USA [Kalúz et al. 2005]. The model calculates the immissions concentrations for determined area in nodal points for the network with chosen step in presented reference points arbitrarily situated in the map of interested area. The model is able to project the range – isolines of arbitrary concentrations according to interpolant of the values of calculated concentration network. In our case they are point values in NE-SW transect of interested area with the centre of the source place. Additional output is numeric calculation of concentration values in determined area. WinModim calculate dispersion of pollution materials on the base of input data about the source of pollutant materials and about dispersion conditions, wind course and speed, air stability and other data.

Precision verification of mathematic model of SO_2 dispersion with consequential transfer on deposit speed will be realised by direct measurement of passive deposit of sorptive-cummulative method. The method determined deposit speed (areal fallout in $mg \cdot m^{-2} \cdot d^{-1}$), by recalculation of volume concentration in $\mu g \cdot m^{-3}$ according to regression equations or graphs presented in ČSN 03 8204, 03 8211 and ST SEV 5292-85, respectively ČSN 03 8203, ST SEV 991-78 [Čeľovský 1997].

RESULTS AND DISCUSSION

The trends of influencing by production recession, introduction of technologies without waste, reconstruction of filtration systems and technological changes of production processes are important arguments in this favourable evaluation. World globalization of production and consumption in process of reallocation of the sources, production and wastes materialized in the market with everything determining the trends for each one in emission market. Paradox of the most up-to-date economics by disobedience to world calls and agreements with the privilege of developing countries on emission of pollutants (or its sale) causes deterioration of the hope for survival of the next generations. The reason of deterioration not striking, but functioning influence is not lonely effect, but environmental deterioration of outliving sort including man. According to the thesis

everything coheres with everything, we will meet with sometimes casual consequences, activities more frequently in interest of extensive development of unappreciated consequences of environmental deterioration.

The example of these tendencies is parallel climatic change. The change is the consequence, but also the factor of the next development. The trend of next development is necessary to consider. Unconsequentionably the climatic change objectively increases ecosystem sensibility for next unfavourable influences, in case of immisions markedly increases the range of critical loads, that means it shifts them to lower values, which were in case of suitable conditional state accepted. It is expected the fact, that alone climatic change as the consequent of untamaebleness immission situation and load will have bigger influence on out living as an actual emission of pollutants. In all cases present situation requires next systematic studies and every one at least knowledge directing to improve present situation is excessively appreciated.

We considered the method precision, equipment availability and service costs during the evaluation of direct measurement methods. The most suitable direct measurement method of sulphur dioxide is sorption-cummulative method according to ČSN 03 8211 (and method elaborated by SVÚDM-Praha, Běchovice). Presented method is determined for long term measurement of SO_2 concentrations and its compounds in the area, where is monitored the degree of aggressive atmosphere influence. The final value of sorpted amount of SO_2 (including sorpted elements of SO_3, H_2SO_4 and soluble sulphates) for areal unit of strong alkalized surface of filters presents immediate aggressive atmospheric influence. The filters are after exposition (30 days) analytic elaborated and by measuring analyses determined the content of sulphates by using of ethanol dilution of barium perchlorate and blending indicator of thorine and methylene blue to pink coloured dilution.

The amounts of detected sulphur compounds in area of AMS LELES during monitored period are presented in the table 1. Up to date there were realised eleven 30 days measuring periods and in time range from 29th Febr.2012 to 24th January 2013. The samples were taken away also after the period of 24th January 2013 up to present time, but the Department of Chemistry has the problem with elaboration of the results.

Table 1. SO_2 concentrations during monitored period – S-K method

Measuring period	Expiration period 30 days	SO_2 concentrations, $\mu g \cdot m^{-3}$	SO_2 amounts (AMS), $\mu g \cdot m^{-3}$
1.	29.02.2012–30.03.2012	25.8	2.7
2.	30.03.2012–29.04.2012	16.06	1.7
3.	29.04.2012–30.05.2012	16.81	1.2
4.	30.05.2012–29.06.2012	33.94	0.6
5.	29.06.2012–29.07.2012	66.28	1.9
6.	29.07.2012–28.08.2012	0.00	3.6
7.	28.08.2012–27.09.2012	0.00	3
8.	27.09.2012–27.10.2012	57.02	2.2
9.	27.10.2012–26.11.2012	32.45	2.8
10.	26.11.2012–25.12.2012	42.87	5.5
11.	25.12.2012–24.01.2013	44.36	3

Measured partial results show bigger occurrence between the individual methods. Though both methods found and recovered increase of SO_2 amount in measuring period from 26th November 2012–25th December 2012 and also decrease of SO_2 amounts in measuring period from 30th March–29th April 2012 and from 29th April 2012–30th May 2012 of our experiment.

S-K- method detects not only the amount of SO_2 but also other compounds as for example SO_3, $SO_{4\text{-}2}$ or H_2SO_4. Real differences found by individual methods will be relevantly judged after long term investigation and analysing of given methods by statistic method of correlation.

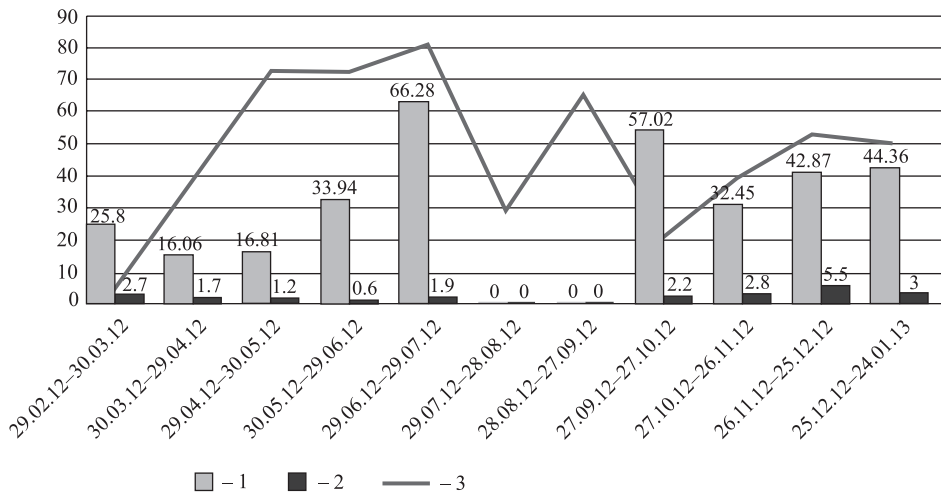

Fig. 2. Total rainfall with AMS and concentration of SO_2 $\mu g \cdot m^{-3}$
1 – Sorptive-cummulative method, $\mu g \cdot m^{-3}$, 2 – AMS method, $\mu g \cdot m^{-3}$, 3 – Rainfalls, mm

The figure 2 shows the development of immission situation after rainfall. The situation in the air is better after rainfal in case of both methods. More detailed explanation of the graph illustration of rainfall total in mm orange line and measured concentrations of SO_2 in $\mu g \cdot m^{-3}$ according to both direct measuring methods. The first type of measurement is on the principle of ultra violet fluorescent method AMS and the second one is measured according to sorptive-cummulative method.

Dispersion maps of immission share of SO_2 ($\mu g \cdot m^{-3}$) for the year 2011 presented on Figure 3 were elaborated on the base of the amount of air emission from the sources SE-EVO and measured values of pollution materials (year average) from AMS Leles in programme MODIM (programme for mathematic modelling of pollution materials dispersion – air immissions). Presented immission share is used for calculation of economic damage caused by immissions in agricultural production.

The calculation of concentrations of SO_2 (NO_x) was realised in model area close to emission source SO_2-SE, a.s. – z. EVO (Power station Vojany). Total sulphur deposit for areal unit was determined.

Determined ratios of the individual sulphur forms in the area were used (SO_2, SO^{4+}, H_2S), the period of abidance in primary form and their rate to total extent of the sulphur in the air. It was determined the total sulphur load, which can be comparing with critical sulphur deposition – the highest sulphur deposition causing vegetation damage (without contribution of nitrogen, respectively in combination with its deposit).

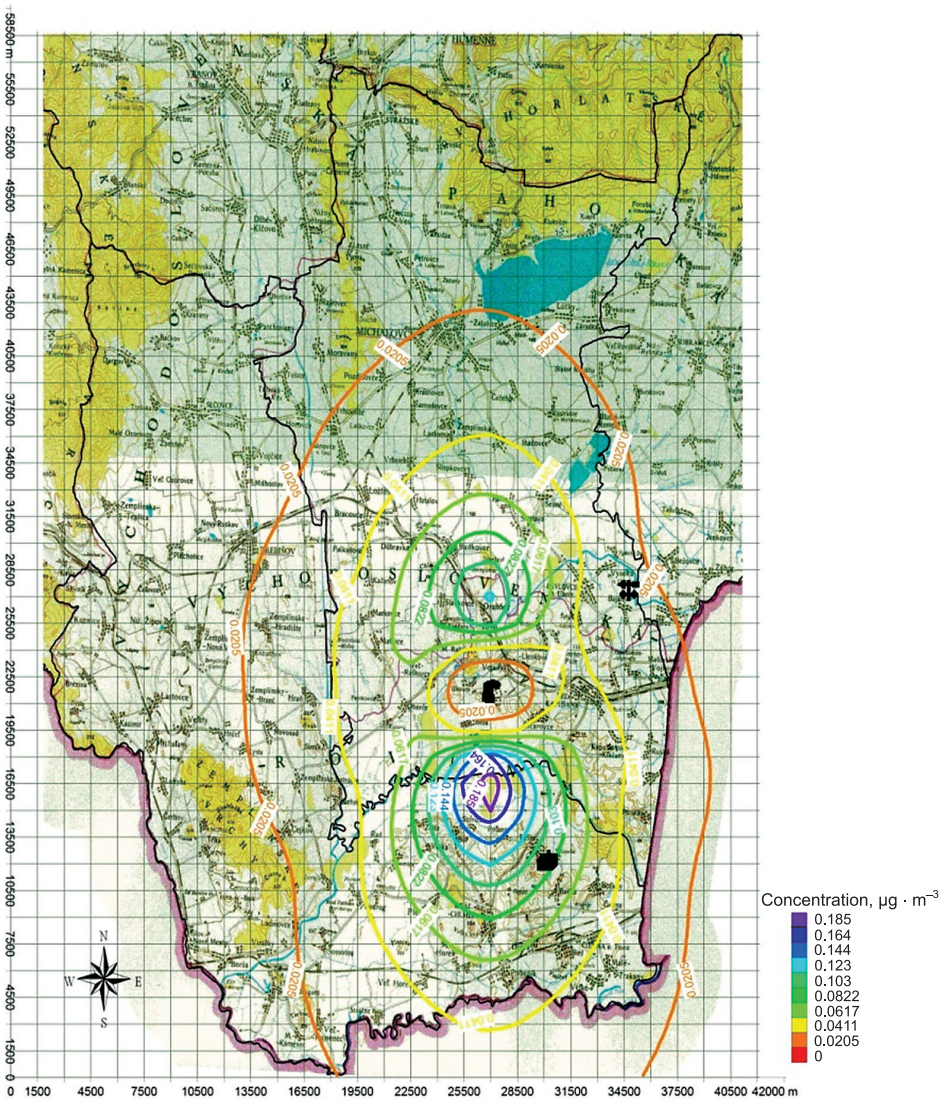

Fig. 3. Range map of immission share SO_2 (µg · m^{-3}) EVO- the year 2012

CONCLUSION

The significant assignment in process of immission harmful effects is monitoring of the air pollution. The methods of measurement of harmful materials in the air are direct and indirect. Determinations of harmful materials concentrations by known methods are financially and operatively very demanding and they are connected with energetic networks.

Presented work monitors SO_2 content in the air, because their portion in the process of air pollution is the biggest. The goal is to determine sulphur territory loading by transformation of SO_2 concentrations on deposit speed according to chosen methods. The work compares two direct methods, sorption-cummulation and chemiluminiscence methods.

The amounts of SO_2 determined by sorption-cummulation method in the individual periods were several times higher as the amounts measured by chemiluminiscence method (AMS Leles). It is caused due to sorption-cummulation method detects not only the amount of sulphur dioxide, but also other compounds as SO_3, SO^{4-2}, or H_2SO_4.

Immission load of given area is expressed by isolones of SO_2 concentrations expressed in $\mu g \cdot m^{-3}$. It was used for this purpose mathematic model of pollution materials dispersion in the ait Win Modim version 4.11 elaborated by ENVItech s.r.o. Trenčín company on the base of ISC2 EPA USA.

Presented work is the first experiment to solve abuse of the agricultural production by immissions in specific area of Vojany loaded by excessive sulphur input from the biggest source of sulphur dioxide in Slovakia. At the same time it is the indication of investigation methodology of given problem by using of smart computer manageable methods. Apparently lighter way with more knowledge will be (in comparison of nitrogen) investigation of sulphur problem as acidification factor of the place of agricultural crops. This assumption confirms the fact, that in wet deposition 60–70% of acidification effects are caused by sulphates in comparison with 30–40% influence of nitrates. Even though the load is not possible to reduce only on acidifaction – acidification caused only by the sulphur and nitrogen, this fact is preferentially important.

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WERYFIKACJA POŚREDNIEJ OCENY OBCIĄŻENIA ŚRODOWISKA ZANIECZYSZCZENIAMI ANTROPOGENICZNYMI

Streszczenie. Ocena obciążeń obszaru wynikających z zanieczyszczeń antropogenicznych stanowi istotny wskaźnik w procesie określenia udziału imisji w zmniejszaniu plonów rolniczych. Pomimo gwałtownej redukcji wszelkiego rodzaju zanieczyszczeń powietrza przeważa opinia, że imisja wciąż występuje, choć w formie ukrytej, i to na skalę istotną ekonomicznie. Dowiedzenie rozmiarów tej skali jest trudne zarówno pod kątem ekonomicznym, jak i technicznym. Dlatego opracowywane są metody pośrednie oceny stopnia zanieczyszczenia, jednak poziom ich wiarygodności wykazuje zasadniczą niewydolność. Określenie stężeń substancji zanieczyszczających tymi metodami jest kosztowne i pracochłonne, a także wymaga współpracy z rozbudowaną siecią komputerową. W niniejszej pracy określono zawartość SO_2 w atmosferze, gdyż związek ten ma największy udział w zanieczyszczaniu powietrza. W celu wykonania pomiarów wykorzystano wybrane metody, które określiły przedział stężeń siarki i azotu oraz poziom stężenia SO_2 i tempo jego usuwania, by wyeliminować kosztowne i pracochłonne metody używane dotychczas. W niniejszej pracy porównano dwie metody bezpośrednie: sorpcyjno-kumulacyjną oraz chemiluminescencyjną. Ilość SO_2 stwierdzona za pomocą metody sorpcyjno-kumulacyjnej w pojedynczych okresach była kilkakrotnie większa od ilości zmierzonej przez stację AMS Leles. Wyniki obrazują relatywnie duże różnice między obiema metodami pomiarowymi. Powyższe metody mogą zastąpić kosztowne i pracochłonne metody intensywne.

Słowa kluczowe: zanieczyszczenie powietrza, imisja, metody pomiaru dwutlenku siarki

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