

Research Article

Overview of Air Pollution Assessment in Northern Europe (Lithuania) by Passive Diffusion Sampling

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The regional air pollution study in Lithuania provided a comprehensive overview of air quality in Lithuania (in Vilnius (capital) and rest of territory) when 375 monitoring sites at different representative locations (urban, suburban, and residential) were equipped with diffusion samplers. The samples were analyzed for sulfur dioxide (SO₂) and nitrogen dioxide (NO₂) concentration. The measurement results show that the mean concentrations of SO₂ in all investigation sites during the study period did not exceed the annual limit value of $20.0 \,\mu \text{g} \cdot \text{m}^{-3}$ and were below the lower assessment threshold value of $8.0 \,\mu \text{g} \cdot \text{m}^{-3}$. The mean concentrations of NO₂ in Vilnius agglomeration exceeded the annual limit value of $40 \,\mu \text{g} \cdot \text{m}^{-3}$ at seven sites and in zone-at three sites with the intensive traffic flow, located near to highway. Comparison of SO₂ and NO₂ concentration levels was performed for 2004-2005 and 2010-2011. The level of nitrogen dioxide concentrations has decreased by 34, 26, 24, and 49% during the next six years in the city of Vilnius, and the concentration of SO₂ in the air environment decreased by 40–60%.

1. Introduction

During the past 20 years, there has been a marked improvement of the air in Europe [1]. As SO_2 produced by burning of fossil fuels significantly contributes to acid deposition, it affects ecosystems and is harmful for human health. Nitrogen oxides are mostly produced during combustion by industrial facilities and the road transport sector.

Nowadays, the main goals of monitoring lie in providing useful up-to-date information to the public on pollutant concentrations in ambient air, as well as supporting economical stakeholders and decision makers in air-quality assessment and management. Instruments for air quality may change in complexity and cost. While air pollution is highest in urban zones, the monitoring efforts are typically concentrated in cities, and little sites represent the background level. The financial resources are not equal in different countries, and there are no possibilities to extend monitoring network or upgrade of equipment. The use of passive samplers greatly reduces the cost and the need of long-term measurement programs [2–4]. Personal passive air samplers have been developed and widely used to measure gaseous air pollutants since their introduction in the late 1970s [5, 6].

Monitoring of air pollution in Lithuania is organized by the Environmental Protection Agency. Currently, Lithuanian national air-monitoring network consists of one mobile, fourteen continuously operating urban stations, and three integrated monitoring (IM) stations. European Union (EU) environment law acts and legislation were applied and implemented by the National Environmental Monitoring Program (NEMP).

In this paper, within the framework of "Lithuanian Air Monitoring System Modernization Using Diffusive Samplers" (LAQMO) project for the first time were evaluated the concentrations of SO₂ and NO₂ by determining the ambient concentration using the passive sampling method at 375 sites in Lithuania. The spatial maps of compounds using geographical information systems (GIS) were evaluated on one year measurements with diffusive air samplers.

2. Methodology

The analysis of concentration for SO_2 and NO_2 using diffusive samplers were set up in the urban background (residential), semiurban (mixed residential and commercial), and roadside (busy street/road and crossing) sites in order to get spatial variation in pollutants concentrations. The obtained data were compared with the acceptable levels of air pollutants that are adopted in the EU as the limit values (Table 1).

2.1. Campaigns. The most appropriate sites for placement were determined. For purposes of taking into account the influence of weather conditions, a network of 375 passive samplers was deployed for all four seasons: autumn (September–November) 2010, winter (December–February), spring (March–May), and summer (June–August) of 2011 and were covered in 8 measurement periods (Table 2).

10% of the sampler was in duplicates, i.e., some colocated passive samplers were deployed at the sampling sites with available continuous monitors for cross correlation and calibration purposes [7]. This information was used for uncertainty calculation in the framework of GUM (Guide to the expression of Uncertainty in Measurement), applied in the laboratory of Passam Ltd., Switzerland (Table 3).

Eight sampling campaigns of 14 days were carried out in Vilnius agglomeration and zone (the rest part of Lithuania). The locations of the monitoring sites in Vilnius and zone selected for the passive sampling is shown in Figure 1.

2.2. Description of Samplers and Measurement Uncertainty. Passive samplers deployed in different city sites were collected after 14 days of exposure time intervals. The passive samplers were provided and analyzed by PASSAM AG (Switzerland). As supplied by the firm, the tubes are protected from sunlight by an opaque cylindrical box. These samples have been exposed to sites with different sources of atmospheric emission and environments (Section 2.1).

Although several approaches to uncertainty evaluation exist, the indirect approach of GUM published by the ISO was used (Table 4). The permanent verification of the sampling rate, based on weight losses of permeation tubes, is an independent way of checking the overall performance of diffusive sampling systems. The output information is important for assessing measurement uncertainty. With this procedure, the requirements of ISO 9001 (process control) was fulfilled as well. Furthermore, with this procedure, long-term stability of results was guaranteed, and measurement results were comparable over time. The calculation of uncertainty started on the basis of the following measurement equation:

$$C_{\rm u} = \frac{\left(m_{\rm d} - m_{\rm b}\right) \cdot 10^6}{\mathrm{SR} \cdot t},\tag{1}$$

where C_u is the ambient concentration, $\mu g \cdot m^{-3}$; m_d is the mass of desorbed analyte, μg ; m_b is the blank of analyte, μg ; SR is the diffusive uptake rate, ml/min; and *t* is the exposure time. The input quantities and their uncertainties are defined as follows:

TABLE 1: Atmospheric air quality (μ g/m³) guidelines for selected air pollutants aiming to protect human health adopted by the European Union Council Directive 2008/50/EB.

	SO ₂	NO_2
Appual limit value (LV)	20	40 (human
Annual minit value (LV)	(vegetation)	health)
Upper assessment threshold (UAT)	12	32
Lower assessment threshold (LAT)	8	26

TABLE 2: The measurement periods.

Season	Period	From	То
I autumn	1	5.11.2010	17.11.2010
	2	17.11.2010	1.12.2010
II winter	3	6.1.2011	20.1.2011
	4	20.1.2011	3.2.2011
III spring	5	25.2.2011	8.4.2011
	6	8.4.2011	22.4.2011
IV summer	7	6.6.2011	20.6.2011
	8	20.6.2011	4.7.2011

TABLE 3: Uncertainty in measurements.

Analyte		Limit value (μ g/m ³)	Standard deviation
NO ₂	Annual mean	40	3.9
	UAT	32	4.8
	LAT	26	5.9
SO ₂	Annual mean	20	5.4
	UAT	12	8.9
	LAT	8	13.4

 u_{m_d} : uncertainty of the mass of absorbed analyte. The standard uncertainty can be characterized by the standard deviation of the calibration function.

 u_{m_b} : blank values. The variation of the blank value has to be added to u_{m_d} in absolute terms $u_{SR}u_{SR}$ —uncertainty of sampling rate. The variation of this term is given by the standard deviation of repeated verification experiments in standard atmospheres.

 u_t : exposure time. This term is in general negligible at exposure times of more than one week. At shorter times, this term has been taken into account.

An additional term has been introduced, which covers the uncertainties budgets of repeated measurements, microenvironmental factors, variations in the geometry of samplers, etc.

 $u_{\rm p}$: variation of multiple samples at the same site. The size of this term is estimated by the median of triplicate samplers in the field.

 u_{ext} : external influences such as temperature, wind speed, and humidity. This term has to be taken into account, if the samplers are used in extreme conditions.



FIGURE 1: Location of diffusive samplers.

TABLE 4: Uncertainty estimation according to GUM.

Component		Limit value ($\mu g m^{-3}$)	Uncertainty combined	Uncertainty expanded	$U_{\rm mean}$
	Annual mean	40	10.8	21.6	7.7
NO ₂	UAT	32	10.2	9.9	7.3
	LAT	26	20.5	19.7	7
SO ₂	Annual mean	20	11	22.1	7.8
	UAT	12	13.1	26.2	9.3
	LAT	8	16.7	33.5	11.9
C ₆ H ₆	Annual mean	5	14.1	28.2	10
	UAT	3.5	17.3	34.6	14.1
	LAT	2	26.6	53.3	18.9

The combined uncertainty u_k is calculated as follows:

$$u_{k} = \sqrt{u_{\text{SR}}^{2} + \left(u_{m_{d}}^{2} + u_{m_{b}}^{2}\right)} + u_{p}^{2} + u_{t}^{2} + u_{\text{ext}}^{2}.$$
 (2)

The expanded uncertainty is calculated by using a coverage factor of 2:

$$u_{k} = 2 \cdot \sqrt{u_{\text{SR}}^{2} + \left(u_{m_{d}}^{2} + u_{m_{b}}^{2}\right)} + u_{p}^{2} + u_{t}^{2} + u_{\text{ext}}^{2}.$$
 (3)

The uncertainty of the mean of the 8 periods is calculated as follows:

$$U_{\rm mean} = \frac{U_{\rm single}}{\sqrt{8}}.$$
 (4)

2.3. Spatial Interpolation. Maps of the pollutant concentrations over the area were obtained by interpolation of the passive sampler measurements. By using custom-made automated scripts on open source GRASS GIS software (version 6.4), the following geostatistical methods commonly used for surface interpolation from randomly



FIGURE 2: Seasonal variation of mean SO₂ concentrations for the entire study period from 3 November 2010 to 4 July 2011 (bar lines show $\pm 22.1\%$ expanded uncertainty).



FIGURE 3: Seasonal variation of mean SO₂ concentrations for the entire study period.

sampled points: inverse distance squared weighting (IDW; GRASS function v.surf.idw), bicubic spline interpolation (BCS; GRASS function v.surf.bspline), and kriging interpolation with automated calibration of parameters (AK; GRASS function v.krige) were tested [8].

By comparing statistical variability of the interpolated datasets, it became obvious that with increasing search radius (N of neighboring points used in interpolation), the IDW method produced rather unstable results, the BCS method under similar conditions (increasing length of splines) produced clearly predictable results with a slight tendency of statistical "smoothing" of the interpolated grid, while AK indicated the most stable statistical results due to its ability to autocorrelate all measurements in the sample [8].

In order to streamline the process of geostatistical data analysis and operational mapping, a customized Linux shell script was developed. It uses geostatistical and mapping functions of the open source GRASS GIS software (v.surf.bspline),



FIGURE 4: Annual mean concentrations of SO₂ in Vilnius.

as well as some of the Linux OS libraries (libgdal, libgeotiff, libpng, etc.) to automatically generate geostatistical grids and operational maps by iterating over each of the polygon objects (urban areas, etc.) by using standard samples of coordinated measurement points as an input. Geostatistical grids will be created in GRASS GIS environment with 10 m pixel size in the standard LKS94 CRS and masked with boundaries of the urban areas. They will be exported from GRASS database as Float64 data type rasters in GeoTIF file format without any associated color table [8].

3. Results

3.1. Vilnius Agglomeration

3.1.1. Sulfur Dioxide. The SO_2 passive samplers were exposed for periods of 2 weeks each at a time over the study

period (120 samples). The values of passive samplers for SO₂ ranged between approximately 0.7 and $1.8 \,\mu g \cdot m^{-3}$. The exceedance above the annual limit value ($20.0 \,\mu g \cdot m^{-3}$) for the ecosystems was not observed. The examination of seasonal variation patterns revealed valuable information. As expected, SO₂ values show seasonal variation. The period of measurement was analyzed corresponding to the four seasons: winter, spring, summer, and fall. The temporal variation for all of the 14 sampling sites is presented in Figures 2 and 3. SO₂ concentration during the entire period of observation ranged from a minimum ($0.15 \,\mu g \cdot m^{-3}$) in summer to a maximum ($3.05 \,\mu g \cdot m^{-3}$) level in winter.

During fall, the mean SO₂ concentration had the highest level (up to $1.80 \,\mu \text{g} \cdot \text{m}^{-3}$) at sites located in the residential and recreation areas with a mean concentrations ranging from 0.5 to $1.3 \,\mu \text{g} \cdot \text{m}^{-3}$. In wintertime, SO₂ concentrations ranged from 0.5 to $4.1 \,\mu \text{g} \cdot \text{m}^{-3}$. The highest mean values for this



FIGURE 5: Seasonal variation of mean nitrogen dioxide concentrations at site-specific areas for the entire study period from 3 November 2010 to 4 July 2011 (bar lines show ±21.6% expanded uncertainty). (a) Vilnius, transport. (b) Vilnius, residential. (c) Vilnius, suburban.



FIGURE 6: Annual mean concentrations of NO₂ in Vilnius agglomeration for the period from 3 November 2010 to 4 July 2011.

study period were found to be between 3.1 and $2.3 \,\mu \text{g} \cdot \text{m}^{-3}$. The results indicate that in springtime, SO₂ concentrations ranged from 0.2 to $4.1 \,\mu \text{g} \cdot \text{m}^{-3}$. The minimum mean value $(0.4 \,\mu \text{g} \cdot \text{m}^{-3})$ of SO₂ concentration was measured at a site in a residential area and the maximum $(3.0 \,\mu \text{g} \cdot \text{m}^{-3})$ in trafficinfluenced area. During the summer study period, SO₂ concentrations ranged between 0.2 and $2.1 \,\mu \text{g} \cdot \text{m}^{-3}$.

Data indicate that at sites in the residential and recreation areas, the higher SO_2 levels were recorded in autumn, winter, and spring, when the emissions from energy production are at their highest level (Figure 4).

Conversely, the lowest SO₂ levels were measured in the summer period. Therefore, the seasonal variability of concentrations should be interpreted using existing knowledge on emission and meteorological patterns. In summary, the mean sulfur dioxide concentration in Vilnius ranged from 0.2 to $3.1 \,\mu \text{g} \cdot \text{m}^{-3}$ with an annual mean of $1.1 \,\mu \text{g} \cdot \text{m}^{-3}$.

3.1.2. Nitrogen Dioxide. The obtained data (35 sites) during all the study period revealed that NO2 concentrations varied considerably, which coincides with the other study depending on the distance of the measurement site from main roads caused by the large numbers of vehicles releasing NO_2 [9]. For the entire study period, the mean concentration for the NO₂ ranged between 9.1 and 55.6 μ g m⁻³ (Figures 5 and 6). The NO₂ concentrations demonstrate a large spatial gradient (up to factor of 5), which indicate that road traffic is an important contributor to the NO₂ concentration in urban environment with a mean concentration above the NO2 limit value of $40 \,\mu \text{g m}^{-3}$. However, the mean concentrations of NO₂ at sites with the minor traffic density were close to the upper assessment value of $32 \mu g \cdot m^{-3}$ (Figure 6). At sites in the most visited areas with high density of motor vehicles, the mean concentration of NO₂ ranged between 26.0 and 42.1 μ g·m⁻³. Thus, an exposure to NO₂ concentrations represents a serious



FIGURE 7: Annual mean concentrations of NO₂ in the zone (58 cities) for the period from 3 November 2010 to 4 July 2011.

risk to human health. The data analysis indicates (Figure 5) that the mean NO₂ concentration during 3 November-1 December 2010 ranged from 9.6 to $53.8 \,\mu\text{g·m}^{-3}$ depending on the site environment. The NO2 concentrations measured at almost all traffic sites were higher than those at the residential or urban background sites and ranged between 18.6 and $53.8 \,\mu \text{g} \cdot \text{m}^{-3}$. The limit value of $40 \,\mu \text{g m}^{-3}$ as annual mean concentration of NO₂ was exceeded at 5 sites with intensive traffic flow: Vilnius 13, 18, 20, 27, and 31. The mean concentration of NO₂ reached a value of $46.9 \,\mu \text{g} \cdot \text{m}^{-3}$. The exceedances above the NO₂ limit value $(40 \,\mu \text{g m}^{-3})$ were not observed at sites in the residential area, and they ranged from 13.0 to $29.9 \,\mu \text{g} \cdot \text{m}^{-3}$. The NO₂ concentrations at sites in the recreation and suburban background areas were in the range $9.6-36.6 \,\mu \text{g} \cdot \text{m}^{-3}$ and were below the lower assessment threshold (26 $\mu g {\cdot} m^{-3}).$ In wintertime (6 January-3 February 2011), mean NO₂ levels ranged from 9.6 to 57.4 μ g·m⁻³ (not shown). The highest mean values of NO₂ for this study period achieved or exceeded the limit value of $40 \,\mu \text{g} \cdot \text{m}^{-3}$ at 7 sites in traffic sites. The upper assessment threshold value of $32.0 \,\mu \text{g} \cdot \text{m}^{-3}$ was exceeded at 3 sites in a high traffic area. In spring, mean values of NO₂ varied from 7.8 to $60.1 \,\mu \text{g} \cdot \text{m}^{-3}$ differing from site to site. Remarkably higher NO₂ concentrations with values of 42.8, 43.0, 54.0, 56.5, and $59.3 \,\mu \text{g} \cdot \text{m}^{-3}$ were observed, respectively, at the trafficexposed sites. As can be seen from Figure 5, higher levels of NO2 were measured during summer at some sites in the

residential and recreation areas $(20.8-28.3 \text{ and } 22.7-40.2 \,\mu\text{g}\cdot\text{m}^{-3}$, respectively). As expected, NO₂ concentration was significantly higher in the residential and recreation areas at the sites influenced by transport emissions. Seasonally averaged concentrations of NO₂ were generally higher during winter and spring nearly at all sites. The lowest NO₂ levels were measured in summer (Figure 5).

3.1.3. The Seasonal Variation of Atmospheric Sulfur Dioxide and Nitrogen Dioxide Concentrations in Zone. The mean concentrations of NO₂ in 40 zones' territory sites, during the study period did not exceed the annual limit value of $40.0 \,\mu \text{g} \cdot \text{m}^{-3}$. The spatial distribution of NO₂ concentrations indicates the tendency to be the higher concentrations in the west part of Lithuania. The principal sources of nitrogen dioxide are traffic and to a lesser extent industry and households. High NO₂ levels, combined with other oxidants, have become one of the major air pollution problems in urban areas. For the entire study period (from 6 November 2010 to 4 July 2011), the mean annual concentrations of NO₂ at different sites in the zone were in the range from $3.6 \,\mu \text{g} \cdot \text{m}^{-3}$ to $59.6 \,\mu \text{g} \cdot \text{m}^{-3}$ (Figure 7). Regarding the annual limit value of $40 \,\mu \text{g} \cdot \text{m}^{-3}$, it was exceeded at three sites with high traffic flow in Klaipeda04 (44.6 µg·m⁻³), Klaipeda09 (44.7 μ g·m⁻³), and Klaipeda11 (51.7 μ g·m⁻³). At the sites,



FIGURE 8: Annual mean concentrations of SO₂ in the zone (58 cities) for the period from 3 November 2010 to 4 July 2011.



FIGURE 9: Time series of mean SO₂ concentration.

which were in an area with the relatively intensive traffic flow (Panevezys01 and Siauliai02), annual mean NO₂ concentrations were found to be 27.3 and $33.9 \,\mu \text{g} \cdot \text{m}^{-3}$, respectively. The exceedances above the NO₂ limit values were not observed at sites in the residential or the suburb areas. Data

indicate that the influence of heavy traffic flows reflected on the annual average NO₂ concentrations at sites located near to the highway A1 (Grigiskes01 and Vievis). Annual average NO₂ concentrations were $40.3 \,\mu \text{g} \cdot \text{m}^{-3}$ and $33.5 \,\mu \text{g} \cdot \text{m}^{-3}$. Annual average NO₂ concentrations were between the lower



FIGURE 10: Time series of mean NO₂ concentration.

and the upper assessment threshold values at sites exposed to traffic in urban environment Mazeikiai $(27.4 \,\mu g \cdot m^{-3})$, Kedainiai $(31.7 \,\mu g \cdot m^{-3})$, Telsiai $(28.1 \,\mu g \cdot m^{-3})$, and Taurage $(34.4 \,\mu g \cdot m^{-3})$. At sites Jonava01 $(\mu g \cdot m^{-3})$, Trakai01 $(26.8 \,\mu g \cdot m^{-3})$, Utena01 $(26.7 \,\mu g \cdot m^{-3})$, and Plunge01 $(26.7 \,\mu g \cdot m^{-3})$, annual average NO₂ concentrations were close or slightly exceeded the lower assessment threshold value. The annual average NO₂ concentrations at major sites in the other towns of zone were in the range from 3.6 to $20.0 \,\mu g \cdot m^{-3}$.

The mean concentrations of SO₂ in 40 zones' territory sites during the study period did not exceed the annual limit value of $20.0 \,\mu \text{g} \cdot \text{m}^{-3}$ and were below the lower assessment threshold value of $8.0 \,\mu \text{g} \cdot \text{m}^{-3}$. The spatial distribution of SO₂ concentrations indicates the tendency to be the higher concentrations in west and southwest parts of Lithuania.

As can be seen (Figure 8), during autumn (3 November–1 December 2010), the averaged SO₂ concentration had the highest value of $5.0 \,\mu \text{g} \cdot \text{m}^{-3}$ at site with crossing of streets. In a residential area, the highest values of SO₂ were $5.0 \,\mu \text{g} \cdot \text{m}^{-3}$ and $4.3 \,\mu \text{g} \cdot \text{m}^{-3}$. At the rest sites in zone, average concentrations of SO₂ ranged from 0.20 to $3.5 \,\mu \text{g} \cdot \text{m}^{-3}$. In winter (6 January–3 February 2011), SO₂ concentrations ranged from 0.30 to $5.40 \,\mu \text{g} \cdot \text{m}^{-3}$, from 0.10 to $2.10 \,\mu \text{g} \cdot \text{m}^{-3}$, and from 0.60 to $2.60 \,\mu \text{g} \cdot \text{m}^{-3}$, respectively. Overall, the SO₂ concentration ranged from 0.2 to $4.8 \,\mu \text{g} \cdot \text{m}^{-3}$ at the rest sites in the zone. The results indicate that in the springtime (25 March–22 April 2011), SO₂ mean concentrations ranged from 0.20 to $1.50 \,\mu \text{g} \cdot \text{m}^{-3}$ and from 0.30 to $2.50 \,\mu \text{g} \cdot \text{m}^{-3}$.

During the summer (6 June-4 July 2011), SO₂ concentrations ranged between 0.20 and $2.10 \,\mu \text{g} \cdot \text{m}^{-3}$. Data

indicated that higher SO_2 levels were measured during autumn, winter, and spring at sites in the residential and recreation areas when the emissions from energy production and heating are at their highest level. Conversely, the lowest SO_2 levels were measured in summer.

3.2. Comparison of SO₂ and NO₂ Concentration Levels for 2004-2005 and 2010-2011. The results of the 2004-2005 and 2010-2011 campaign in Figure 9 show that in Vilnius, the level of sulfur dioxide concentrations in the five years has not changed significantly. Significant decrease in SO₂ concentrations was observed at 03 and 05 in Klaipėda sites located in residential areas, while in the cities of Kedainiai and Palanga, the concentration of SO₂ in the air environment decreased by 40–60%.

The level of nitrogen dioxide concentrations has decreased by 34, 26, 24, and 49% during five years in the city of Vilnius at the sites next to traffic. Also the increase of NO_2 concentration was observed at Žirnių street and at the crossroads of V. Kudirkos Street near Pamenkalnis (Figure 10).

4. Conclusion

Concentrations of SO₂ and NO₂ were determined over a year using the passive sampling method. For the entire study period (from 3 November 2010 to 4 July 2011), the annual mean concentrations of SO2 ranged between 0.20 and $3.40 \,\mu \text{g} \cdot \text{m}^{-3}$ in 40 zones territory sites. The SO₂ annual averages were below the value of $1.50 \,\mu \text{g} \cdot \text{m}^{-3}$ at all sampling sites (except two). These values demonstrate rather small differences and the even regional pollution by SO₂ and its strong connection to the long-range transport of SO₂ on the regional scale. The emission of SO_2 from the local sources more or less formed the level of pollution at those sites. Mean concentrations of NO₂ ranged between 2.3 and $9.4 \,\mu \text{g} \cdot \text{m}^{-3}$ in 40 zones territory sites. The annual mean concentrations of NO₂ were in the range $3.0-5.0 \,\mu \text{g} \cdot \text{m}^{-3}$ at the sites in major part of the territory and were significantly below the lower assessment threshold limit value of $26.0 \,\mu \text{g} \cdot \text{m}^{-3}$ for the annual NO₂ concentration. The highest annual average concentrations of NO2 were measured at sites close to road with intensive traffic.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the paper.

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