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14

ANALYSIS OF METHODS OF AIR QUALITY ASSESSMENT. APPLICATIONS IN ESTONIA

VELJO KIMMEL

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I Tammet, H., Kimmel, V. and Israelsson S. 2001, Effect of atmospheric electricity on dry deposition of airborne particles from atmosphere Atmospheric Environment 35, 3413-3419

II Kaasik M., Kimmel V. and Kaasik H. 2001, An air quality modelling system for a medium-sized town: a case study in Estonia International Journal of Environment and Pollution. 16 (1-6): 519-527

III Kimmel V., Tammet H. and Truuts T. 2002, Variation of Atmospheric Air Pollution in Conditions of Rapid Economic Change – Estonia 1994 - 1999. Atmospheric Environment 36, (25), 4133-4144

IV Kimmel V., Kaasik M. 2002, Assessment of Urban Air Quality in South Estonia by Simple Measures. Environmental Modelling and Assessment (accepted)

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MAIN ARGUMENTS PROPOSED

1) The study of Estonian monitoring data reveals that spatial coverage and the list of air pollutants monitored requires enhancement.
2) Air quality on Estonia satisfied the Estonian and European standards with few exceptions during 1994 – 2001. Most remarkable exception was high level of ozone concentration in remote areas.
3) A simplified CORINAIR scheme developed for the calculation of emissions from mobile sources can be used for a harmonised correction of official emissions in the past calculated without the splitting of vehicles into classes.
4) Atmospheric electric field enhances the dry deposition of fine particles on the top branches of trees. Gamma-spectroscopic determination of radon daughters attached on fine particles or on cluster ions settled on the needles/leaves of the trees was developed as a method for quantitative proof of the effect. Strong effect of electrical deposition was found on the trees under the HV power lines.
5) Simple measures like passive monitoring, Gaussian-plume modelling of pollution dispersion, traffic counts, and additional inventories of pollution sources form a gentle basis for air quality assessment in medium-sized cities like Tartu.
6) The optimisation of Air Quality Assessment requires a holistic analysis of all aspects associated with pollution creation-transport-deposition. These aspects can be covered by emission inventories, the modelling of meteorological fields and pollution distribution and the analysis of the data of monitoring and scientific studies.

INTRODUCTION

Air quality assessment with all its aspects is an important way to understand the status of the environment. Warning of people, and the forced reduction of traffic and business activity in case of violations of environmental standards are becoming a reality in many cities. Progress in telemetric systems, computer software and communication technology have made environmental data more readily available for everyone. The integrated approach towards environmental management implemented in the developed countries is well in line with the concept of sustainable development. However, keeping the advanced systems operational requires a holistic analysis of all relevant data to determine why, what and how to monitor.

Air quality is one of the most critical environmental issues in urban settlements, influencing people's health and accelerating degradation of materials. Therefore, international agreements have been laid down in many documents like European Union Directives (COM, 1996, 1999), and the WHO quality guidelines (WHO, 1987, 2000). These papers have been based on the results of scientific research and are to be supplemented with new knowledge.

The growing flux of raw information about the levels of air pollution and the emissions of pollutants increases the amount of work that is required to comprehend and appreciate the environmental data and frightens the decision makers – the planners and the authorities. Thus, the development of the methods of data analysis and the simplification of decision-making process appear as the essential tasks in the progress of environmental monitoring and management.

General methods of air quality assessment are

1) monitoring of concentrations of air pollutants at the sites where polluted air affects people, plants and materials;
2) monitoring of emission and deposition of pollutants;
3) modelling of emission, distribution and deposition of air pollutants, and estimating of pollutant levels according to the emissions and the relations between pollutants.

The selection and/or development of a certain method necessary for air quality assessment depends on the air pollution situation and economic opportunities on site. Thus, every country, region and city can perform assessment according to their own methods considering the objectiveness and completeness of the process. The aim of the current work is to evaluate the already used methods of air quality assessment, and if necessary, to develop new ones for application in a small developing country.

The basics of air pollutant concentration monitoring, the description of the realization of the monitoring system, and the analysis of monitoring results in Estonia are the subject of Chapter 1 of the Thesis.

The basics of the calculation of air pollutant emissions and the situation of pollutant emissions observable in Estonia are given in Chapter 2. The analysis is based on an international standard CORINAIR established for inventorying pollution sources. Chapter 2 presents a simplified scheme for the calculation of emissions from mobile sources in the conditions of limited information about the properties of vehicle classes. The results of the simplified scheme are compared with the results of more sophisticated schemes and a good agreement is found. An evaluation of the shares of different emission sources of Estonia is given in the last section of the chapter.

Air pollution modelling - the third general method of air quality assessment is the subject of Chapter 3. The main factors of pollution fields are local emissions, the transport of pollution from remote sources and meteorological parameters. In the thesis, the validation of modelling results is made in two areas - air quality modelling and the prediction or simulation of atmospheric deposition. The predictions of air quality by the model AEROPOL are checked by passive monitoring of NO_2 and SO_2 levels in Tartu and North-East Estonia. Agreement between the modeled and measured values is fair. Up to now the theory of the deposition of pollution has not corresponded well with the observed phenomenon that prevalently, the top branches of trees are damaged by air pollution. It has been found that this phenomenon could be explained by the electrostatic deposition of air pollutants on the tips of needles and leaves where the atmospheric electric field is efficiently concentrated.

The methods for the assessment of air quality (AQA) are described and practical examples (including optimisation of AQA) are presented in Chapter 4. The scheme developed for AQA in Tartu is a low-cost version of complex assessment programs running on-line in developed countries. Chapter proposes also ways to use developed methods in optimization of air quality assessment. The chapter summarises the results of the previous chapters.

The thesis deals with classical pollutants – sulphur- and nitrogen oxides, particulate matter, ozone and carbon monoxide. However, the ideology worked out is easily applicable also for the assessment and monitoring of other pollutants, such as heavy metals, hydrocarbons, etc.

The thesis includes references, acknowledgements and summaries in English and Estonian. The attached publications discuss some aspects of the study in detail: the influence of electric field on dry deposition of particles (Tammet, Kimmel, Israelsson, 2001), urban air pollution modelling and validation of modelling results (Kaasik, Kimmel, Kaasik, 2001), the analysis of the data of Estonian air pollution (Kimmel, Tammet, Truuts, 2002), and the study of air quality assessment by simple measures (Kimmel, Kaasik, 2002).

1. AIR QUALITY MONITORING

1.1 Aims of Air Quality Monitoring

The aims of modern Air Quality Monitoring (AQM) are the prevention of human health and ecological systems, particularly the Earth as a whole, from harms caused by air pollution, and the creation of databases describing the state of environment. There are also many other scientific or health-related purposes for performing monitoring activities in the atmospheric air. The tools for achieving these aim(s) are: 1) monitoring of air status in the frame of integrated environmental monitoring, 2) air pollution prediction and control, 3) inventorying of pollution sources, 4) addressing the consequences of air pollution by the estimation of percentages of people, ecosystems, cultural objects etc. suffering from air pollution, and 5) the reporting of data-findings in easily readable form.

The air quality monitoring is a part of the environmental management system and therefore closely connected to other branches of the management system. The AQM is connected with everyday economic activities by their air pollution production characteristics. Emission calculations and measurements' results are included into the same frame for pollution level predictions in a modern AQM system. The restrictions or limiting some human activities should follow in case of high pollution level(s).

The AQM is the only way to get information about the actual levels of pollutants in an acceptable, reliable and proper manner. Thus, a set of rules has been agreed upon internationally to carry it out in a comparable and good-quality way.

1.2 Monitoring methods, standards, quality

The modern Air Quality Monitoring System consists of on-line measuring devices; a data management system including data transfer and quality control systems, and databases; models for predicting pollutant levels; data presentation tools - GIS, the Internet, and the decision support system. The output of the system -- reliable data, is at the same time a tool for decision-makers or planners.

The list of exact methods used in AQM depends on the specified objectives and available resources. The main classes of methods used concern measurement, modelling and reporting. The selection of methods depends rather on the availability of finances and the level of institutions involved in monitoring. The characteristics of various measurement method types used in AQM have been discussed by Sivertsen (1997) and WHO (2000) - see Table 1.1 At the same time the measurement methods determine also the opportunities for validation the model's and presenting the data.

The time and space scale of the monitored processes determine the type of measurement instrumentation. For example, in deposition monitoring it is sufficient to use daily average concentrations. Thus sequential, or even sometimes passive monitoring sufficiently fulfils the requirements.

Table 1.1 List of instrument types for air quality monitoring and their characteristics.

Instrument type	Type of data collected	Data availability	Typical averaging time	Typical price (US $)
Passive sampler	Manual, in situ	After lab analyses	1-30 days	10
Sequential sampler	Manual/ semi-automatic, in situ	After lab analyses	24 h	1 000
Monitors	Automatic. Continuous, in situ	Directly, on-line	1 h	> 10 000
Remote monitoring	Automatic. Continuous, path integrated (space)	Directly, on-line	<1 min	> 100 000

The development of passive samplers in the last decades has given a cheap and reliable measurement tool which can be used in monitoring. Good examples of passive monitors implementations have been described by Bower *et al*. (1991), Svanberg *et al*. (1998). Recommendations for using passive samplers in monitoring activities have been given in many reports of global authorities (GAW, 1997; WHO, 2000). Such methods are especially important in the developing countries with limited resources, such as Estonia.

The compatibility of recorded environmental data is achieved by using standard methodologies and requirements. For measurements it is achieved by using internationally approved methods and procedures (CEC, 1999, 2000), and Quality Assurance (QA) procedures (Lalas and Saeger, 1998). The list of standard methods and limit values are presented in Table 1.2. The target of data quality requirements is the time coverage and accuracy of measurements. The time coverage of continuous measurements should exceed 90%, and, consequently, of indicative measurements 14% (evenly distributed over the year). The accuracy of measurements required depends on pollutants - for SO2 and nitrogen oxides 15% and 25% are applicable, for particulate matter and lead, 25% and 50%, respectively for continuous and indicative measurements.

The obligations of countries to report and publish information obtained through monitoring are in Europe fixed by Directives and decisions (COM, 1996, 1997, 1999, 2000, 2001). The collected information is often readily available also on the Internet. The development of computers and associated software and the implementation of the Internet and geographical information systems (GIS) in reporting have made the information easily accessible and

well understandable. Thus, the decision makers and the public have better opportunities to use information collected in monitoring.

Table 1.2 Standard methods of monitoring and the limit values of pollutants.

Pollutant	standard method in urban areas	standard method in remote areas	limit values ($\mu g\ m^{-3}$) - averaging time
sulphur dioxide	UV-fluorescence - ISO 10498	impregnated filter + Ion Chromatography	350 - 1 hour 125 - 24 hours 20 - 1 year
nitrogen oxides	Chemi-luminescence ISO 7996	impregnated filter + photo-colorimeter	200 - 1 hour 40 - 1 year 30 - 1 year
carbon monoxide	IR absorption	-	10 mg m^{-3} - 8 hour
PM10	weighing EN 12341		50 - 24 hour 40 - 1 year
ozone - O_3	UV absorption ISO 13964	UV absorption ISO 13964	200 - 1 hour 110 - 8 hour 65 - 24 hour

Sources: COM (1992, 1999, 2000), EMEP (1996)

1.3 Air Quality Monitoring in Estonia

1.3.1 Aims and structure

The air quality monitoring system in Estonia was created in the Soviet time and some parts of it are still in operation. The established air pollutants limit values have been stricter than in the western countries, but nobody cared when pollution levels exceeded the limit values. Monitoring stations in cities were usually located for urban background measurements. The monitoring was performed mainly by manual sequential sampling methods and only few continuous monitors of Russian origins for CO measurements were running. In the remote areas, two stations were measuring within the EMEP network. No inter-calibration activities were held between laboratories and the overall quality of the data is usually unknown.

The aims of Estonian AQM, which have been defined within the framework of overall environmental monitoring, are to identify and fix trends in air pollution and pollution load levels. Locations of operational stations, the traffic network and the biggest polluters are presented in Figure 1. The most polluted region is North Estonia, where locate the bigger cities and heavier polluters, such as power plants, the Kunda cement factory, oil shale mining and chemical industries.

Today the Air Quality Monitoring of Estonia has 3 stations in remote areas, 3 stations in Tallinn, and 16 precipitation stations - see Figure 1. A background research station is situated at Tahkuse, where also ions and UV radiation are measured as described by Hôrrak *et al*. (1998), and lichenoindication sites in about 100 places where the deposition of heavy metals is measured every fifth year. Air pollution measurements are accompanied by meteorological measurements at EMEP stations at Vilsandi, Lahemaa and Tahkuse.

Figure 1.1 Map of Estonia with continuous air monitoring stations, monitored parameters in 2001, main cities and major pollution sources

An aspect hindering the work of air quality monitoring is the lack of monitors of particulate matter - see discussion in Kimmel, Tammet, Truuts (2002). Fortunately, PM10 monitors operate at stationary stations since autumn 2000, and county centers are visited by a mobile van equipped with all instruments under contracts to carry out temporary measurements. Results obtained at county centers are, due to a short time coverage, suitable only for objective estimation.

A weakness of the monitoring program established in remote areas is clearly visible in Table 1.3. The measured parameters do not allow to calculate the total deposition of acidifying N- and S-compounds, which is one of the main aims of EMEP program. Thus, the data of collected at the stations cannot be fully exploited for further studies.

Table 1.3 EMEP measurement program 2000 (Estonian share in bold)

	Component	**Measurement period**	**Measurement frequency**
Gas	**SO_2, NO_2**	**24 h (at Vilsandi 1 h)**	**daily**
	O_3	**hourly values stored**	**continuously**
	Light hydrocarbons C_2-C_7	10-15 m	twice a week
	Ketons and aldehyds (VOC)	8 h	twice a week
	Hg	24 h	weekly
Particles	**SO_4^{2-} (only at Lahemaa)**	**24 h**	**daily**
	Cd, Pb (1. priority), Cu, Zn, As, Cr, Ni (2. priority)	week	weekly
Gas (g)+ Particles (a)	HNO_3(g)+NO_3^-(a), NH_3(g)+NH_4^-(a)	24 h	daily
	POPs (PAH, PCB, HCB, chlordane, lindane, A-HCH, DDT/DDE)	to be decided	to be decided

Precipitation	**Amount, SO_4^{2-},NO_3^-. pH, NH_4^+, Na^+, Mg^{2+}, Ca^{2+}, K^+, conductivity**	**24 h**/weekly	**daily** or weekly
	Hg, Cd, Pb (1st priority), Cu, Zn, As, Cr, Ni (2. priority)	**weekly**	**weekly**
	POPs (PAH, PCB, HCB, chlordane, lindane, A-HCH, DDT/DDE)	to be decided	to be decided
Measurements of VOC, heavy metals and POP are made only at few sites.			

Source: EMEP, http

1.3.2 Monitoring development in 1990-2000

At the end of the 1980s, the Estonian AQM consisted of 17 urban stations – Tallinn (8), Kohtla-Järve (4), Narva (2), Narva-Jõesuu (1), Kiviõli (1) and Jõhvi (1). Manual collection was carried out 4 to 6 times a day and wet chemical methods were used at all stations. The pollutants monitored include mainly classical pollutants with some specific hydrocarbons as tracers of production typical to specific sites. In the mid-1990s air quality monitoring was started also in the second largest city Tartu, as well as in Pärnu, using the same routine. Continuous monitors were installed at one station in Tallinn only in 1994 and later in other stations – see Table III in the supplementary publication 4.

Two remote area stations included in the EMEP network operated from the end of the 1970s in Lahemaa and Sõrve. The Sõrve station was transferred to Vilsandi in 1994; one additional integrated monitoring site with air quality monitors started operating at Saarejärve in 1997.

Round-year precipitation samples were collected on 5 locations mainly in the northern part of the country in the early 1990s. 1999 saw an expansion and samples were collected on 16 sites. Pollution loads to surface were determined from the chemical composition of snow all over Estonia in winters with good snow coverage. The number of samples taken varied from 40 to 220 in different years and enabled to create a pollution load map for various pollutants (Roots *et al*., 1998). Thus the spatial coverage of the monitoring of pollution load was significantly enhanced.

The list of pollutants monitored saw the same kind of enhancement: when in 1994 only 6 continuous monitors were in operation, already 12 operate in 1997, and 22 in 2001. The last significant expansion occurred due to the help of the Phare Air Accession Program which gave the complete set of 6 stations (monitors of SO_2, NOx, O_3, CO, PM10), a van with a rack for monitors and a meteorological mast, and advanced software for monitor calibrations, data transfer, storage and validation.

Some changes were made in the list of stations - Majaka station was closed in 1999 and a new Õismäe station was opened in the North-West residential area of Tallinn in the beginning of 2001. All stations are now equipped with continuous monitors of nitrogen oxides, SO_2, PM10, CO, O_3. Suspended particulate matter (SPM) is measured additionally at Viru traffic station. Continuous monitors are now operational also in Kohtla-Järve (since May

2002), and Narva. There are plans to restart monitoring in Tartu, which was stopped in the end of 2000, the main constraints are financial.

1.3.3 Main trends in pollution levels in the 1990s

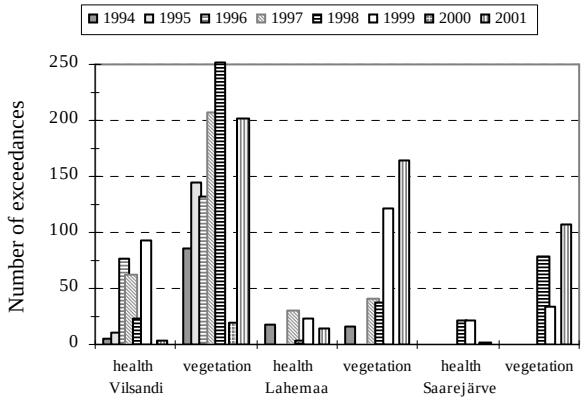
The description of main trends is divided into two parts according to the availability of data and, thus, the applicability of the analysis to all Estonia.

1.3.3.1 Gaseous pollutants

The detailed analysis of data for different pollutants measured at EUROAIRNET stations of Viru, Rahu, Majaka, Lahemaa, Vilsandi, Saarejärve is given by Kimmel et al (2002). Here, the time period considered is extended by two years until the end of 2001.

In general, the study reveals that the short-term concentrations of pollutants in the capital city of Tallinn are low due to moderate climate and the location of stations near the sea, causing windy weather. The levels of ozone, often exceeding the limit values, are the most dangerous aspects - see Figure 1.2. The exceeding of the limit values is mostly associated with vegetation protection limits. However, health related limits are also exceeded in suburban stations and in remote areas.

The levels of sulphur dioxide and carbon monoxide are much lower than the limit values. The levels of nitrogen oxides are quite stable despite increasing traffic loads. This phenomenon can reflect the fact that the replacement of old cars with high emission rates by newer cars with low emission rates has been most intensive in capital. The trends mentioned by Kimmel et al. (2002) are almost same, but even more pronounced for NO_2 and CO - see Figure 1.3. In the recent years a clear annual cycle with maximums in spring is observable for NO_2.



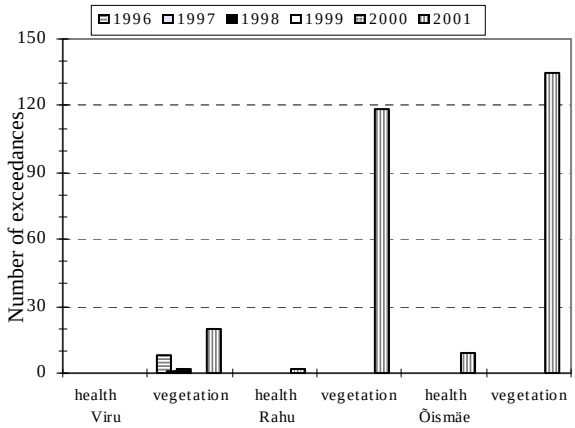


Figure 1.2 Number of violations of limit values of ozone in remote area (upper) and urban area (lower) figure.

The analysis of pollution levels at background stations shows that these levels have been elevated in Vilsandi that is located closer to bigger polluters in Central and Eastern Europe and to international shipping.

The average concentration of nitrogen dioxide could be close to the yearly limit value also in other South Estonian county centers Tartu (about 100, 000 inhabitants) and Viljandi (about 22, 000 inhabitants) (Kimmel, 1998). In these towns the levels of pollutants were elevated due to a limited spread of pollution because of a river valley (Tartu) or narrow streets (Viljandi). The evaluation of hourly maximums from two-week average concentrations measured by passive monitors is given in subchapter 4.2.2.1. Air quality monitoring is thus seen to be important also in other cities of Estonia.

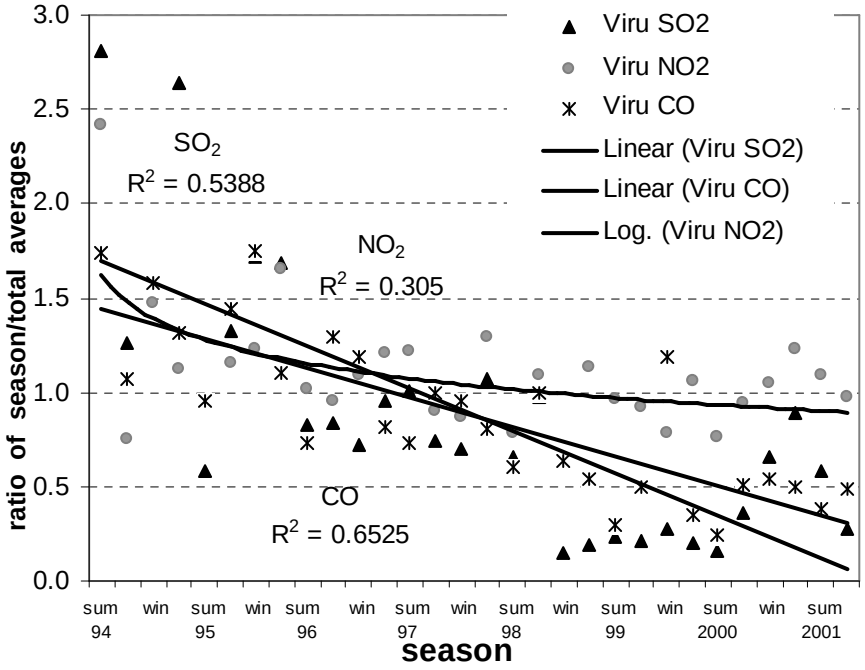


Figure 1.3 Variation of pollutant concentrations at urban station Viru 1994-2001.

The analysis of North East Estonian monitoring data supports the outlines drawn earlier. According EERC (1999) and Tihane, Vazova (2002) in case for NO_2 the limit values were exceeded in 3 cities – Kohtla-Järve, Kiviôli and Narva, and for some specific hydrocarbons such as aldehyds and phenols, and hydrogen sulphide near chemical enterprises and purification plants from 1994 -2002. Since wet chemical methods were used with intermittent sampling 4-6 times a day for half an hour each, and the quality of data is unknown, the collected data are suitable only for objective estimation.

The analysis of Tartu monitoring data given in the supplementary article 3 (Kimmel, Kaasik, 2002) shows that pollution levels increased in the most parts of the town during the 1990s.

1.3.3.2 Particulate matter

The values of particulate matter monitored from 1. May 1995 to 31. December 2001 at the Viru traffic station exceeded the Estonian limit value (150 µg m^{-3} as 24-hour average) 134 times in 19.9 % of cases. Since the time coverage of monitoring was too short for proper assessment the outline given is only indicative.

Seasonal variation of the concentrations of particulate matter at the Viru station in center of Tallinn is expressed on Figure 1.4. The highest spring

values mentioned in the supplementary article 4 are still there and the limit values are frequently violated.

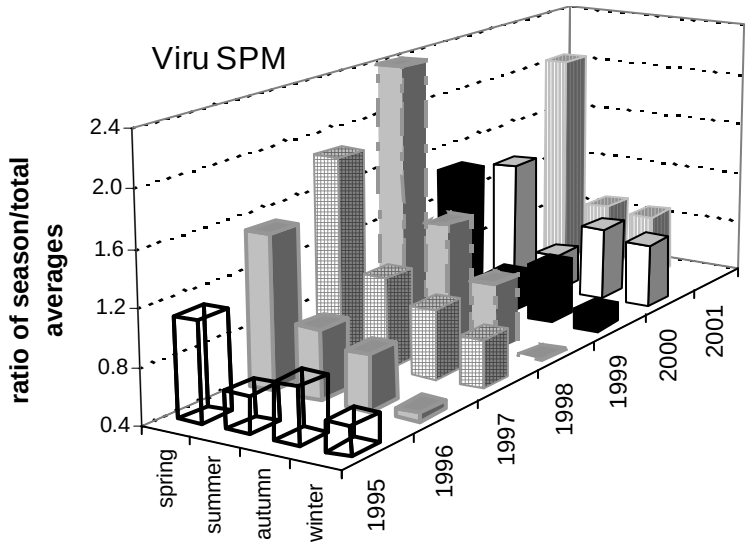


Figure 1.4 Seasonal variation of particulate matter concentrations at Viru station in 1995-2001.

The particulate matter levels monitored at 3 sites around the Kunda cement factory showed a significant drop from 1994 to 2000: maximum dropped 2.5 times and yearly averages 2.6 - 7.9 times.

Monitoring of particulate matter (SPM) by intermittent sampling in North - East Estonia in the 1990s shows that the limits were exceeded in 4 towns - Sillamäe, Kiviôli, Narva and Narva-Jôesuu. In the last two, the violation of standards was probably associated with pollution from power plants – see Figure 1.

Monitoring of lead concentrations since 1998 shows that the levels are considerably below the limit values -maximum concentration is 0.15 µg m^{-3}, verifying the low usage of leaded gasoline in transport.

One of the aims of AQM is to prevent or alert to damage to human health. Little is known about direct health risk of total suspended particulate matter SPM, but there is plenty of information about health risks associated with smaller particles with diameters less than 10 µg PM10, less than 2.5 µg PM2.5, or even smaller. Thus, I try to estimate from the collected SPM values the figures of the concentrations of smaller particles in atmospheric air.

Estimation of PM10 values

Evaluation of PM10 concentration from the monitored SPM values is made using relationships found in parallel measurements of SPM and PM10 by beta-attenuation (EERC, 2001). The analysis of data reveals that the ratio of PM10

to SPM decreases significantly when the SPM values exceed several hundred micrograms per cubic meter. The analysis shows also that the best fit is obtained when we distinguish the heating-period from other periods of time that express the different origin of particulate matter in different seasons discussed by Kimmel, Tammet and Truuts (2002).

Thus, the estimation of the PM10 levels was made with the fitting function PM10 = SPM × (0.0004 × SPM - 0.038) +26.578 for the heating period and the function PM10 = SPM × (-0.0006 × SPM - 0.3414) +1.418 for non-heating period, which represents the measured values with an accuracy expressed by a standard deviation of 0.755 in the heating period and of 0.496 in the non-heating period. The average value of the estimated PM10 - 27.3 µg m^{-3} was close to the EU upper assessment threshold of 30 µg m^{-3}. The obtained 98% percentile was 73.6 µg m^{-3}, thus the potential to exceed the limit value is estimated to be high.

The estimation is in good agreement with the findings in Central and Eastern Europe expressed by Houthuijs *et al.* (2001) that PM10 and PM2.5 values are much higher in the named region than in US or Canada and form a severe potential danger to human health. The agreement with real measurements is also quite good - in 2001 the average value of PM10 monitored at Viru was 30 µg m^{-3}, and 98% percentile equals to 94 µg m^{-3} (EERC, 2002).

Thus, the chapter points out some pollutants requiring enhancement of monitoring and gives some ideas where is reasonable to perform air quality measurements.

2. EMISSIONS OF AIR POLLUTION

The exact amount of air emissions is impossible to measure, and therefore, some calculations must be used (Milford and Russell 1993). The commonest approach is to divide the sources into groups, thereafter measure one characteristic source in a group and assume that other sources in the group emit the same amount of pollutants. The grouping of sources and the commonest emission rates within the groups are internationally standardised. The standard is named CORINAIR (Co-ordination INformation Environmental and AIR Emission Inventory) and if filled correctly, gives an enhanced summary of pollution in a compatible form. For many areas the standardisation is not completed and thus different calculation schemes are used.

2.1 International standard CORINAIR

CORINAIR splits emission sources into 11 main sectors (see Table 2.1), 76 sub-sectors and 375 activities.

Table 2.1. Main sectors in CORINAIR94 with examples and their relation to economic sectors as defined in the Fifth Environmental action Programme

Category	Examples	Target sectors
I Combustion in Energy and Transformation industries	Power stations, fuel (coke, gas, nuclear town gas) production.	Energy
II Non-industrial Combustion plants	Agricultural, domestic combustion, public services, railway stationary sources	Energy and Consumers
III Combustion in manufacturing Industry	Cement, glass, metals and lime production.	Industry
IV Production Processes	Chemical, food, asphalt, textile industries.	Industry
V Extraction and Distribution of fossil fuels	Pipes, petrol distribution.	Energy
VI Solvent and other product Use	Aerosols, tyre manufacturing.	Several
VII Road transport.	Buses, cars, other road vehicles.	Transport
VIII Other Mobile sources and Machinery	Agricultural power units, aircraft, railway, shipping, fishing, domestic machinery.	Transport
IX Waste Treatment and Disposal	Landfills, lightning, incineration, batteries, sewage sludge disposal.	Several
X Agriculture	Agricultural soils, pesticides, livestock, field burning.	Agriculture.
XI Other	Forests, Natural Fires.	-

The emission load from stationary sources can be checked by measurements. In case of mobile sources, there is only an indirect way for checking the emission calculation results - by measuring the formed deposition loads and pollution fields.

The summing of the emissions of a country, region or a settlement is a compilation work, since it uses results of research groups dealing with calculations of emissions from the sources hard to evaluate, such as forests (Isidorov, 1999), agricultural areas, and human dwellings (NAEI, 2002).

The quality of emission data is characterised by an uncertainty factor giving an estimation of the deviation of the calculated values from real emissions. The dependence of the figures of uncertainty factors on the quality of the calculation schemes used, omitted from Aalst et al. (1999), are given in Table 2.2.

Table 2.2 Correspondence of uncertainty factors and procedures used for the calculation of air emissions.

rating	definition	typical error ranges
A	an estimate based on a large number of measurements made at a large number of facilities that fully represent the sector;	± 10 to 30 %
B	an estimate based on a large number of measurements made at a large number of facilities that represent a large part of the sector;	± 20 to 60 %
C	an estimate based on a number of measurements made at a small number of representative facilities, or an engineering judgement based on a number of relevant facts;	± 50 to 150 %
D	an estimate based on a single measurements, or an engineering calculation derived from a number of relevant facts and some assumptions;	± 100 to 300 %
E	an estimate based on an engineering calculation derived from assumptions only;	± order of magnitude

Source: EMEP/CORINAIR Guidebook

2.2 The scheme for calculating emissions from mobile sources

The author developed a simplified CORINAIR scheme for calculating the emissions from mobile sources. Below, the calculation schemes for emissions from mobile sources are described in more detail.

Two schemes are available for calculating emissions from mobile sources. One of them is based on fuel consumption data and the other on traffic activity data – see equations 2 and 3. The selection of one or the other scheme depends on the availability and the quality of data and on the pollutant considered. The emission of sulphur dioxide, carbon dioxide and lead from vehicles does not

depend on driving mode. Thus, an assumption is made that all sulphur, carbon and lead in gasoline are combusted (oxidised) and emitted into the atmosphere. So, only fuel consumption data can be used for emission calculations. The amount of other pollutants emitted by traffic, namely, nitrogen oxides, carbon monoxide, hydrocarbons and particulate matter depends heavily on the driving mode. Therefore, the selection of an exact model depends rather on the time and space scale of the issue under investigation.

$$Q_i = m_i \sum_j p_{ij} \frac{t_j l_j}{Tl} \qquad (2)$$

$$Q_i = T \sum_j q_{ji} \frac{t_j}{T} \qquad (3)$$

where

i indicates the pollutant,

Q the total emission of the considered pollutant

p_{ij} the emission factor of the considered pollutant based on fuel consumption for the j-th vehicle class and driving mode (kg per ton)

q_{ij} the emission factor of the i-th pollutant based on the driven distance for the j-th vehicle class and driving mode (kg per km)

l the specific fuel consumption, averaged over all vehicle classes and driving modes (l per km)

l_j the specific fuel consumption for the j-th vehicle class and driving mode (litres per km)

T the total traffic activity (vehicle-km/a)

t_j the traffic activity for the j-th vehicle class and driving mode (km/)

On the local scale, the calculation is normally based on the traffic intensity data. On the regional or global level, when the total emission of a country or some region of a country is considered, both schemes give satisfactory results. The selection of one or the other scheme depends, therefore, more on the accuracy of the basic data. According to Larssen (1989), for diesel-powered engines, the fuel-based emission factors are considerably more stable, that is, they vary much less according to vehicle class and driving mode, than the activity-based emission factors. The same seems to be true also for the accuracy of statistical data for fuel consumption, which are usually more accurate than traffic activity data.

2.2.1 International standard CORINAIR

The calculation of emissions from mobile sources is based on CORINAIR standard for driven kilometres. Accordingly, vehicles are divided into different classes, such as passenger cars, trucks, bikes, etc. Thereafter, the driven kilometres of a certain vehicle type and driving mode are multiplied by the

emission rates (grams of emitted pollutant per kilometre of drive) and the amount of emissions from a certain vehicle class are found.

2.2.2 Methods used in Estonia

In Estonia the methodology for the calculation of emissions from mobile sources has not yet been well established. The Russian methodology without splitting vehicles into classes, methodologies of the Nordic countries and combined methodologies have all been used. Several studies have been performed relying on the established European CORINAIR methodology (Mäkela and Salo, 1994; Kimmel, 1998a; COWI, 2000), but officially published data about emissions from mobile sources is scarse. Table 2.4 gives the main characteristics of used methodologies.

2.2.3 Simplified CORINAIR scheme developed

Due to the fact that some data required for calculations by CORINAIR scheme are absent in the official statistics of Estonia, a slightly different scheme was developed by the author (Kimmel, 1998). The deviation from standard is a basis for the emission calculations for fuel consumption data, not for driven kilometres as required by the original CORINAIR scheme.

There are two reasons for such deviation - the accuracy and availability of statistical data, and the including of agricultural vehicles into calculations. Until the end of the 1990s no statistics was available about the driven kilometres of different vehicle classes in Estonia. The accuracy of the assumed driven kilometres by different vehicles is believed to be much more inaccurate than the figures of fuel consumption of different vehicle classes. This assumption is supported by two facts. First, the studies dealing with finding the number of kilometres driven by different vehicle classes in Estonia in the mid-1990s were made only in city conditions and therefore do not cover the whole of the country. Second, the data of annual total fuel consumption are easily available. Third, the emission factors per fuel consumed depend much less on the vehicle and environmental conditions, such as the age of the vehicle or air temperature than the emission factors given per driven kilometre (Larssen, 1989).

The values of emission rates used in the developed schemes for calculating emissions from mobile sources are given in Table 2.3. The figures are based on papers by Larssen (1989, 1993), Mäkela and Salo (1994), Oehme et al. (1991), Alexepoulos and Assimapoulos (1993) and Giess (1998). To calculate the given factor it was assumed that the driving mode fell into urban (30%) and rural (70%) divisions. Since there were only a few highways in Estonia in the middle of the 1990s, their role was evaluated to be negligible and therefore not taken into account. This aspect must be taken into account when using the scheme for

calculating the emissions of a later period. On highways the emission rates for NOx should increase and for CO and VOC decrease

Table 2.3 Pollutant emission factors (g/kg of burned fuel) for motor vehicles.

Pollutant	Passenger cars	Trains	Tractors	Trucks	Buses	Motor-cycles
NO_x	30	20	50	50	60	5.5
VOC	25	40	100	70	20	50
PM_{10}	1.6	5	3.5	3.5	2.5	2
CO	200	50	300	150	45	300

The next Table 2.4 gives the figures for emissions from mobile sources calculated by different methodologies from 1990 to 1997. The values in the first column for years 1993 and 1996 differ from those in paper Kimmel (1998) due to corrections in the official amounts of consumed fuel.

The table shows that a simple scheme developed for calculating emissions from mobile sources gives reasonable results in comparison with more advanced schemes the results of which are given in the third column. The official figures seems to be overestimated for CO.

Table 2.4 Comparison of methodologies for calculating air emissions from mobile sources of Estonia (kilotons per annum).

Methodology	Kimmel (1998)			Official values			Other studies		
Basis of calculation	fuel consumption			fuel consumption			driven kilometres		
Splitting of vehicles into classes	yes			no			yes		
	NOx	CO	VOC	NOx	CO	VOC	NOx	CO	VOC
1990	53.1	187	79	45.1	374	70.4			
1992	28.7	103	46	24.5	175	34.4	17.8	76.5	23.5
1993	30.6	107	44	27.4	189	37.3			
1996	27.9	95.1	39.2	29.2	238	44.6			
1997	29.1	97.7	40.9	28.1	256	47.6	31.3	74.6	20.6

Sources: Kimmel (1998), Mäkela, Salo (1994), COWI (2000)

The accuracy of values in Table 2.4 depends significantly on the accuracy of the values they are based on – COWI (2000) states that the uncertainty of fuel consumption data could be as high as 50% in some years. Obviously, the

uncertainty of the calculated emission figures can not be much lower. Another important factor influencing uncertainty is that all emission factors were taken from the studies made in other countries and no studies were made to reveal the applicability of the found emission rates in Estonia. Thus, the uncertainty of all given figures is quite high.

2.3 Data of air emissions of Estonia.

2.3.1 Economy changes in the 1990s

Air pollution is mainly determined by human activity and depends on economic development. Estonia is a small country (area of 45227 km^2 and population of about 1.5 millions), which was occupied by the Soviet Union until 1991. Restoration of the independent state initiated rapid economic changes resulting in the onset or disappearance of different air pollution problems. The growing number of cars (increase from 1990 to 1999 of almost 90% (SOE, 2000)) and the drop in agricultural activities to almost one half influence the patterns of air emissions and consequently, the air pollution levels.

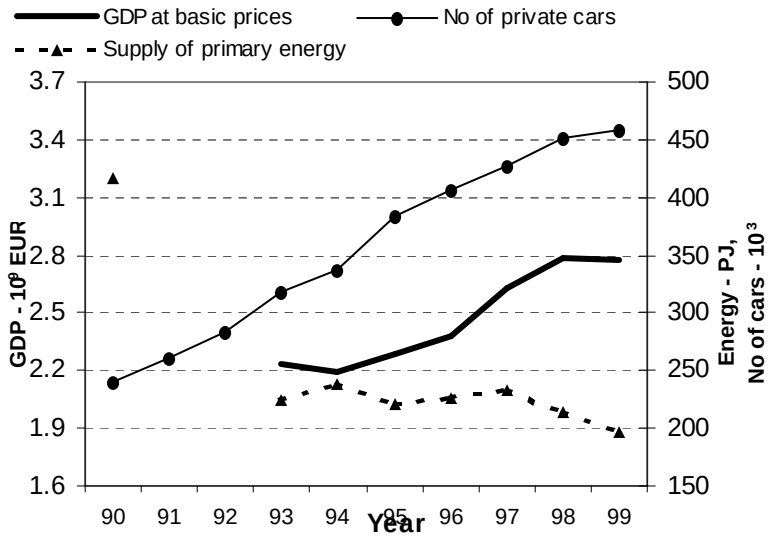


Figure 2.1 Characteristics of economic development of Estonia in 1990-2000.

The main outlines of economic changes are given in the supplementary article 4 and presented in Figure 2.1 and Table 2.5. During the period from 1993 to 1999 Estonian GDP grew 23%, export grew 3.3 times and import 4.3 times, expressing the restructuring of state-owned economy into private one, and renewal of production equipment.

Reconstruction of economy and reduction of electricity export to Russia and Latvia by several times resulted in a drop in electricity production from 17181 GWh in 1990 to 8521 GWh in 1998 (SOE, 1999). The new economical system forced energy conservation. As a result, heat production decreased 2.3 times during the same period of 1990-1998 (SOE, 1999).

Table 2.5 Basic economy data of Estonia in the 1990s.

Year	Export 10^9 DEM	Import 10^9 DEM	Production of meat –kilotons of slaughter weight	Sown area of field crops (km^2)
1990			182.5	11163
1991			177.1	11143
1992	0.69	0.64	131.6	11066
1993	1.33	1.48	83.7	10570
1994	2.12	2.69	69.4	9350
1995	2.38	3.41	67.7	8507
1996	2.66	4.37	58.6	8591
1997	3.65	6.12	53.4	8642
1998	4.41	6.90	60.0	8611
1999	4.43	6.31	61.1	8187
2000	6.73	9.03	52.7	8098

Sources: SOE (2000)

2.3.2 Official published data of air emissions

The availability of data on air emissions in Estonia depends on source types. Emissions from stationary sources are calculated quarterly on routine basis and reported in yearbooks published since 1993. The emission data is collected in two stages. First, the county environmental offices (altogether 20) collect the initial data. Subsequently the Environmental Information Centre of the Estonian Ministry of Environment checks the primary data, calculates the totals and produces yearly public reports.

The official data about air emissions during the last 10 years is presented in Table 2.6. The variations shown in the Table are mainly caused by the reduction of many industrial activities, including oil shale mining (compared with the figures for 1990, mining had been reduced almost by half by 1999), and decrease in electricity and heat production as stated in previous sections. The Kunda cement factory and two big power plants offer good examples of progress in reducing air emissions from big local polluters. The amount of particulate matter emitted from the Kunda cement factory dropped from 61.1 kilotons in 1993 to 31.4 kilotons in 1995 and 0.5 kilotons in 1999 (Roots, 1996; Kohv and Kirjanen, 2000). Similar drop can be observed also in the emissions from two power plant – 129 and 54 kilotons of particulate matter were emitted in 1990, only 34 and 34 kilotons in 1995 and 24.7 and 35.8 kilotons in 1999 (Kohv and Kirjanen, 2000). This is a result of economic changes, transition to

EU regulations of air emissions and a dramatic increase in pollution fines controlled by government.

The quality of emission data depends on the first level – county collection. The data collection is supported by the regulation of permissions for emission in the counties. The emission amounts are calculated by the enterprises according to the officially determined rules and presented with an application for the permission to local authorities. The application is carefully checked and approved either without remarks or with some obligatory conditions to the enterprise. The current results are presented quarterly by the enterprises and verified by the county authorities. Thus, an estimate of total emissions accordingly to the permissions given to the enterprises is available before the commencement every year, and the data of actual emissions is available in the end of quarter. In this way, the emission inventory of stationary sources is administered satisfactorily.

Table 2.6 Official figures of air emissions from anthropogenic sources of Estonia (kilotons per annum)

Year	CO Stationary	CO mobile	NO_X Stationary	NO_x mobile	SO_2 Stationary	SO_2 mobile	VOC Stationary	VOC mobile	Particulates Stationary
1990	59.9	374.2	22.6	45.1	238.8	13.1	18.0	70.4	268.5
1991	56.8	342.4	20.9	42.4	232.7	12.7	17.0	64.9	277.8
1992	32.5	175.3	14.9	24.5	179.2	8.2	11.0	34.4	240.8
1993	27.8	182.4	12	26	145.0	8.7	5.7	35.9	189.0
1994	31.8	209.3	14.6	26.5	141.1	8.1	4.7	39,9	161.5
1995	27.2	215.1	14.8	27.2	110.3	8.3	6.5	41	113.1
1996	29.4	238.3	16.3	28.1	117.2	8	5.7	44.6	98.9
1997	26.7	256.1	15.6	29.2	111.0	8	6.3	47.6	78.3
1998	26.4	254.3	14.9	31.1	100.9	9.2	5.7	48	69.8
1999	20.9	194.4	14.4	25.2	94.6	7.9	5.0	37.3	70.5
2000	19		15		92		8		59

Sources: Roots et al. (1996), Kohv and Kirjanen (2000)

The weakness of such administration is the lack of emission measurements. Thus, the calculations of emissions are mainly based on assumptions and engineering calculations. The uncertainty of the given figures, although not published officially, could be close to 100% - see Table 2.2.

2.3.3 Results of scientific studies

Academic research reveals that the official emission figures of nitrogen oxides, carbon monoxide and particulate matter cover only about one half of all the emissions on the local level in Tartu (Kaasik *et al.*, 2001). In smaller towns

the role of official figures is even smaller - in Kärdla, according to GAIAA (2000), they account for less than one third of total emissions. The mentioned studies, representing medium and small towns, enable to evaluate total emissions from domestic heating in Estonia. The role of domestic heating was evaluated by multiplying the figures of the country's stationary emission by factors given for Tartu and Kärdla. The biggest sources (power plants, chemical factors), which are not characteristic for residential areas were priorly subtracted from the total emissions of the country. As we can see, the biggest contribution of domestic heating is observable for CO and particulate matter, reflecting the fact that combustion in smaller heating furnaces is less efficient.

Table 2.7 Emissions from domestic heating of Estonia in 1998 (percentage from stationary sources' emissions)

pollutant	Tartu		Kärdla		Estonia	
	ton/annum	%	ton/annum	%	kton/annum	%
NOx	121	31.3	5.9	114	1.7	11.3
SO2	103	24.9	1.2	33	8.4	8.3
CO	554	29.4	63	61.5	5.8	22.2
PM	1336	209	54	860	23.8	33

Most recently are published also first results of official values of emissions from domestic heating. The values given for the year 2000 are (in kilotons per annum) SO2 – 1.31; NOx – 1.85; CO – 80.62; PM – 16.47 (Kohv et. al 2002). Thus, a good agreement for NOx and particulate matter is observable. The reason for difference could be shortages in statistics (not all firewood is accounted) or difference in consumption of fuels in different years.

2.3.4 Estimation of the share of emission sources of Estonia.

The previous pages present the results of studies performed for emission calculations in Estonia. We can see that there is no data of emissions from biogenic sources – nature and agriculture. Since, according to literature, their role in volatile organic compounds and particulate matter emissions is big, I shall attempt to estimate their emissions in Estonia. In such a way, we get more realistic picture of the shares of different emission sources.

Literature estimates of world-wide emissions of NMVOC are 60-140 million tons from anthropogenic, and 1150 tons (of carbon) from biogenic sources per year (Atkinson, 2000). The share of biogenic sources in the emissions of different countries depends on a climate zone, vegetation cover of the territory and the structure of economy.

In a warm climate, and of lively economy area -California the share of biogenic sources in hydrocarbon emissions does not exceed ten percent (Benjamin *et al*, 1997). In Poland, forests produce 186-763 kilotons of VOCs depending on weather and forming one third to one fourth of all country emissions according to Isidorov (1999). Isidorov used empirical equations and the data of age structure of different trees for his calculations. Separate

calculations were made for the emissions of isoprene and non-isoprene for every common tree species.

The share of biogenic sources in Estonia is estimated to be bigger than in Poland since the anthropogenic emissions of organic compounds are only 40 - 60 kilotons a year, and the standing store of forests is only 3.5 times less. The age of forests determining foliage biomass emitting hydrocarbons is also younger (in the last 60 years forest area has grown for 2.4 and during the recent 40 years for 1.6 times) than in Poland. The structure of Estonian forest differs from that in Poland; coniferous trees account for about half of all forests (instead 78% in Poland). Instead of pine (65.7% vs. 24.9%), birch (5.2% vs. 13.4%) and alder (4.5% vs. 7.6%) are common in Estonia. The role of oak, beech and silver fir forests (together covering 15.5% of the standing store in Poland) is negligible in Estonia.

The exact evaluation of VOC emissions from biogenic sources of Estonia was made by using the ratios of the standing store of forests in Poland and Estonia. The Estonian climate is colder, but the formulas contain absolute temperature, so the difference in results is evaluated not to exceed ten percent. In addition to that, a large amount of organic compounds should originate from mired areas covering about 20% of Estonia land. The influence of the colder climate and other biogenic sources are evaluated to eliminate each other. Thus, an estimation of biogenic emissions of VOC in Estonia was made using the forests scheme described above. The estimation gives us an average of 100 kilotons of VOC per annum. The uncertainty of the given figure could be several-fold, since it is based only on the assumptions made in another country and no supporting measurement results exist.

There has published already first results of calculation of emissions of organic compounds from forest using emission factors found from (EMEP/CORINAIR, 2001). The result of calculation for year 2000 equaled to 34.982 kilotons (Kohv, Mandel, Ljamtsev, 2002). Thus, a fine agreement with estimation made by similarity scheme is observable.

The calculation of natural NMVOC emissions is difficult for most of European countries - in the EMEP database (EMEP, 2001), only 17 countries out of 34 report natural emissions of volatile organic compounds. The biggest values are observable for Russia (5500 kilotons - 60% of total NMVOC emissions), France (427 kilotons - 30% of total) and the United Kingdom (178 kiloton - 10% of total).

There is also a difference between the official results and other studies - Poland reports less than 1 kiloton, while Isidorov (1998) showed that emissions only from forests exceed 100 kiloton. The official values may need some correction also in other countries.

The importance of the emissions of bioorganic volatile compounds is even growing when we consider the potential of organic compounds to form ozone. In England the role of bioorganic sources grows from about 10% in total

emissions of organic compounds to almost 15.4% in the potential to form ozone (NAEI, 2002).

Similarly, one important source of particulate matter is nature: according to Seinfeld and Pandis (1998) nature formed up to 90% of total particulate emissions in the 1980s. The results of the particulate matter monitoring in Estonia in Chapter 1.3.3.2 show that the concentrations rise significantly in spring, which could be a result of natural processes following the melting of snow coverage and anticipating the formation of plant canopy. Thus, the role of natural sources of Estonia is evaluated to be half of the stationary sources' emissions after subtracting the biggest sources (power plants, cement factory).

The summary of official figures and estimated values of emissions are given in Figure 2.2. Figures are presented only for NOx, VOC and PM, since, according to the previous Chapter, they form the main nuisance in Estonia.

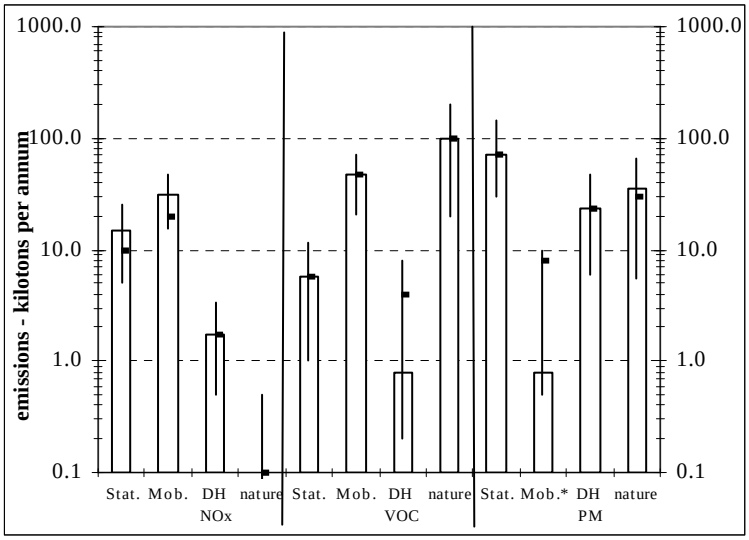


Figure 2.2 The share of different sources in the emissions of pollutants in Estonia (Stat- stationary sources, Mob-mobile sources, DH - domestic heating).
Columns express official or estimated amounts, horizontal line - uncertainty, vertical line - range of confidence.

The Figure expresses firmly that nature seems to be a sector responsible for most of VOC and a large amount of particulate matter emissions. It also reflects the facts that the emissions of particulate matter from road paving and volatile organic compounds from domestic heating have not been studied in Estonia. The uncertainties of emission amounts are high due to too few measurements and/or forgetting some important sources as discussed earlier. In comparison with the uncertainties in the UK emission inventory that not exceed 50%, Estonia has still a long way to go.

Thus, to summarise the findings given in Chapter 2, the database of Estonian sources of air emission requires enhancement.

3. AIR POLLUTION MODELLING

Air pollution is a 3-dimensional phenomenon in nature. Thus, modelling is the only way to obtain knowledge about the spatial distribution of pollution. The accuracy of air pollution modelling depends on the knowledge of input data (meteorology, air emission sources, limiting obstacles) and the processes running in the atmosphere and influencing pollution distribution, deposition, and chemical transformation.

The modelling of air pollution distribution is divided into two big groups – the receptor-oriented and source-oriented models. The receptor-oriented models require the results of pollution levels. Analysis of results called also 'source apportionment study' made in regional (Swietlicki, 1989) or local (urban) scale (Swietlicki et al., 1996) enables to estimate the share of different sources in measured values. The source-oriented models require the data of sources, and the knowledge of processes running in the atmosphere and they allow predict pollution levels at a site. The author work in modelling was related to the validation of results of both types of models.

The accuracy of modelling is set by COM (1999) through the margins of differences with measurements. The exact values of the allowed differences depend on the time period and pollutant considered.

3.1 Parameters affecting pollution field

When air pollution has been created, it starts to transfer in the atmosphere. Obviously, the atmospheric processes affect pollution distribution. Created pollution field is mainly determined by local emissions, pollution transport from remote sources and distribution patterns – meteorological conditions (WHO, 2000). Since all the named aspects determine the formation of pollution fields in almost separate ways, the influence they might have is also characterised separately in the following subchapters.

3.1.1 Local emissions

The description of patterns of air emission has been given in the previous chapter. As we have also seen in developed countries, many gaps exist in determining the amounts of emissions from different sources. Mainly it concerns area sources, small enterprises and sources harder to evaluate, such as domestic heating.

Pollution distribution in the atmosphere is determined by the parameters of emission source. The height of the source is one of the key factors determining the formation of elevated pollution levels. The higher is the source located, the lower are the formed pollution levels on the ground. The pollution level in an urban area is mainly determined by traffic emitting pollution at the height of less than one meter - close to the human breathing zone. The importance of

domestic heating in the formation of elevated pollution levels also comes clearer when we consider that the chimneys of houses emit pollution at the height of about 10 meters - much closer to the ground than the chimneys of boiler houses. Therefore, domestic heating is also one of the coded reasons for the exceedance of the limit values of air pollution as legislated in COM (2002).

3.1.2 Pollution transport from remote sources

An important factor determining the pollution field especially in remote areas is the transport of pollution. Due to chemical transformation in the atmosphere pollution from remote sources is usually more oxidized and therefore differs from the chemical composition of direct emissions. Pollution transport from remote sources has been rather well studied regarding big sources, such as power plants emitting pollution at the height of several hundred meters (Kaasik, Liblik, Kaasik, 1999). Pollution transport has also been well studied with respect to the transport of pollution between bigger areas, such as the whole countries. Such processes are called transboundary fluxes and they have been well studied in EMEP network through the net of modelling -monitoring exercises.

We can take Estonia as an example of changes in transboundary fluxes. The description of the reported figures have been given by Kimmel, Tammet, Truuts (2002) - see the emission part of the supplementary article 4. The drop in the emissions of pollutants mentioned at the end of the 1990s is even more remarkable for the whole decade. In 1998, the emissions of the countries responsible for most of pollution import to Estonia form only 24.3-56.8 % for SO_2, 45-84% for nitrogen oxides, and 44-88% for NMVOC in comparison with the values for 1990. Thus, if throughout the 1990s the patterns of the transport of air masses were similar to what they had been in 1998, the import of pollution to Estonia reduced significantly during the decade.

3.1.3 Meteorological conditions on site

Although the emissions of pollutants determine the concentrations of pollutants, meteorology also plays a substantial role. The most important meteorological conditions are wind direction and speed in both vertical and horizontal directions, and temperature.

The meteorology is usually included into air pollution models. Pielke and Uliasz (1998) and Plate (1999) summarise and discuss their use and influence on air pollution. Authors conclude that modern air pollution models are not using all the power of weather forecast models generating 3-dimensional field of meteorological parameters. Rotach (1999) even shows how different physical description of wind field can result in several-fold differences in the formed pollution field. The main conclusion of mentioned papers is that the accuracy of air pollution models is rather determined by shortcomings in

simulation of meteorological parameters, and not so much influenced from the description of pollution transformation in atmosphere.

The direct effect of temperature is well seen in the conditions of inversion when warmer air masses are higher than colder, the vertical distribution of pollution has stopped and pollution stays in one and the same layer for a long period, forming a health hazard. In urban conditions the buildings, roads and parks form a gentle basis for temperature fluctuations and rise and thus increase the turbulence, which accelerates the distribution of pollution.

3.2 Deposition mechanisms of pollution

When pollution has once been created, it removes from the atmosphere by deposition on the surface or obstacles, and chemical transformation. Deposition process is commonly divided into two parts: dry and wet deposition. Wet deposition means the deposition of pollutants carried by precipitation. Dry deposition means the deposition of pollutants when no precipitation event is observable. In many occasions it is rather difficult to distinguish between wet and dry deposition.

3.2.1 Dry and wet deposition patterns

Wet deposition can be divided into the scavenging of gases or particles by precipitation, cloud interception, fog deposition and snow deposition (Seinfeld, Pandis, 1998). In all the named processes three steps are necessary for wet removal of material – 1) species removal into the condensed water, 2) scavenging by hydrometeors and 3) delivering to the Earth's surface.

Dry deposition is usually characterised by the forces involved in the process. The common processes considered are gravitational, aerodynamic (impaction and interception) and Brownian diffusion (Weseley and Hicks, 2000).

3.2.2 Electric field influence on dry deposition of particles

The previous subchapter shows that electric field is usually neglected in dry deposition mechanisms. However, already in 1977 Wilkening showed that in thunderstorm strongly influenced the concentrations of radon daughters. Study by Henshaw et al (1996) showed that an enhanced deposition of radon daughters occurs near the indoor electric power cables. Measurements by (Tammet, Kimmel 1998) outdoors support this effect. All the named papers explain the enhanced deposition of radon daughters by the effect of charged clusters. Later investigations, explained in detail in supplementary article 1, show that electric field influences also the deposition of charged particles.

The main outcomes of theory developed by (Tammet et al., 2001) shows that for most of particles the influence of electric field on the deposition of particles is stronger than gravitational forces. In comparison with other

deposition mechanisms electric field is especially important for particles with the diameter of 10-200 nm in the conditions of low wind speed (< 2m s^{-1}).

3.3 Validation of modelling results

A model is a simplified description of reality. Every simulation result must be compared with a real situation. The comparison process according to pre-determined rules is called validation and 'model validation kits' consisting from data bases of spatial and temporal variation of pollutants and meteorological parameters created are described by Olesen(1994). The following pages describe the activities performed by the Author in this field.

3.3.1 Validation of modelling results in urban and background areas

The validation of results of air pollution modelling by Gaussian plume model AEROPOL (Kaasik et al., 1999, 2002), successfully validated against international validation kit (Kaasik, 2000) is based on the concept of measuring the pollution field. Mainly NO_2 and SO_2 were considered. Passive monitors were used to reveal long-term average concentrations of pollutants in the atmospheric air. The description of monitors and their validation for NO_2 is given in supplemented article 3, for SO_2 description of samplers is given in Ferm and Svanberg (1998).

The main aim of modelling in a remote area was to study deposition. Thus, passive monitoring and snow samples were selected to validate results. The description of the structure of validation has shortly been given by Gronskei et al. (1998), results have been given in article Kaasik and Sõukand (2000). Generally, the measurements show slightly higher results than predicted by model, which is explainable by absence in model of several source types like transport and smaller point sources, and overestimation of measuring method (see Heal et al., 1999, and supplemented article 3). Additionally, AEROPOL does not include chemical transformation of pollutants in atmosphere. Thus, the lower values of NO_2 by modelling are expectable, since NO_2 is a secondary pollutant formed in the atmosphere from the mainly emitted NO through the oxidation.

The structure of validation in an urban area is described in the supplementary article Kimmel, Kaasik (2002). The difference in the modelled and measured values of pollution was mostly associated with the underestimation of emission rates of pollution sources or the underestimation of the role of chemical transformation of emitted pollutants during transport to the measuring point. The overall accuracy of predictions was smaller than 50%, thus it satisfied perfectly the quality requirements presented by COM (1999).

3.3.2 Validation of the results of simulation of deposition

The validation of the results of the simulation of dry deposition of aerosol particles performed by the author of the thesis started in spring 1997. All work

was performed within the Laboratory of Nuclear Spectroscopy, which is a division of the Institute of Physics in Tartu. Two gamma detectors were used for the studies: HPGe detector GEM-35200 made by EG&G Ortec (later referred to as GEM) and BSI detector BDRG-32355 (later BSI). GEM was supplied with the analyser 92x-W3 Spectrum Master and both detectors with the software package Maestro enabling the identification and quantification of the measured lines of radiation spectra.

The first activities performed for validating the simulation results were associated with the measuring of Pb210 in the samples of plants. The half-life of the named lead isotope (see Table 3.1) enabled sample preparation and the collected samples were dried and measured with BSI detector. During this and the later campaigns, the samples of materials were taken at 3 sites in a suburb of Tartu: a remote area, under and near high voltage (HV) power lines 330 kV. Natural radioactivity, determined mainly by radon daughters, is low in the region– the concentration of ^{226}Ra in soil does not exceed 30 Bq kg^{-1}, being several times less than in North Estonia and ten times less than in the Nordic countries. Plants used for sampling were spruce, deciduous trees and grass. Samples from trees were taken at two heights – at about 2 m and at about 6 m. Samples of tree foliage were taken both inside the tree crown and at the top (or singular branch) of the tree.

The first results of ^{210}Pb measurements and the later measurements of ^{210}Pb in the deposits of different soil layers from the same places did not show any significant difference in the patterns of deposition of ^{210}Pb, which could have been initiated by the effect of electric field on dry deposition of radon daughters. Such a deviation of simulation predictions from the measured values could be explained by the fact that ^{210}Pb had originated from other sources, such as fuel combustion in boiler houses.

Table 3.1 Characteristics of radon daughter elements measured by author.

Isotope	Half-life	Identification energies (keV); its probability (%)	Origin of decay chain
^{210}Pb	22.3 years	46.5 ;	^{222}Rn
^{212}Pb	10.6 hours	238.6; 43.3	^{220}Rn
^{214}Pb	26.8 min	351.6; 35.8	^{222}Rn
^{214}Bi	19.7 min	609.5; 44.8	^{222}Rn

Thereafter, head of the Laboratory Mr. Realo suggested that radon daughter elements with shorter half-life, such as ^{212}Pb, ^{214}Pb, ^{214}Bi should be measured. Thus, attention was transferred to the preparation of samples that would enable to measure the named isotopes. Since these isotopes appear in the pilot and study samples, their characteristics are given in Table 3.1.

Several sampling strategies were analysed and proved. The total number of samples in 1997 included 30 samples of plants and 24 samples of soil. The developed sampling strategy consists of the following main points - 1) samples

of different plants were taken at one and the same site as close to each other as possible; 2) samples at one and the same site were taken repeatedly to enable the elimination of causal variation; 3) samples were taken during dry season thus minimising the disturbing influence of precipitation; 4) samples at different height horizons were taken from one and the same tree; 5) the sampling of grass was performed by taking samples from corners and the centre of a quadrate of about 2x2 m size; 6) in needle samples the share of older and younger needles was almost equal; 7) the collected plant samples were weighted to establish whether the amount was sufficient for measurements; 8) the contamination of samples was minimised by using plastic bags and closing them tightly after sampling.

The developed methodology for sample preparation in laboratory include - 1) the sample preparation in the laboratory was performed within one small room that was cleaned after every preparation; 2) in the laboratory the samples of foliage were quickly cut and inserted into the measuring container; 3) the size of the measuring container was optimised to enable measurements of low concentrations of radioisotopes and also to enable a reasonable sample preparation period. The short time of half-life of measured isotopes required the multiply registration of spectra usually after every hour.

The obtained peaks in the registered spectra of radiation were identified and quantified by standard methodology. In Table 3.2 are presented the main results of a study in 1997. Note, that sometimes, especially for ^{210}Pb, the results are given with uncertainties, which even exceed the measured values. Sometimes, the element was not measurable or the found concentration did not exceed the background concentration.

Table 3.2 Average concentrations (Bq kg^{-1}) of radon daughters in plant samples in different conditions.

	1997			1998	
Isotope	Grass	Spruce 2m	Spruce 6m	Spruce 2m	Spruce 6m
^{210}Pb Remote area	12.4			7.8	18.3
Under HV lines				40.5	11.3
^{212}Pb Remote area		22	42	13	35
Near HV lines					
Under HV lines	155	54	66	10	300
^{214}Pb Remote area			105	60	240
Near HV lines					
Under HV lines	84	345	510	45	510
^{214}Bi Remote area		70	225	215	480
Near HV lines					
Under HV lines	140	140	420	120	1350

The analysis of results reveals that the biggest difference in radioactivities of vegetation was observable for spruce. Consequently, in later measurements in 1998 only spruce samples were taken. The maximum values measured in

1997 were equal to several hundred decays per kilogram of sample – in radiation units Bq per kg in spruce needles under HV lines. It was also observed that in the remote area the radioactivity of needles on a jutting off branch was close to the values observed under power lines. Thus, the effect of electric field on the deposition of charged ions-particles was measurable even in natural conditions.

The obtained experience in sampling and in the preparation of samples allowed to repeat measurements in a slightly modified way in 1998. The most significant change was that sampling was carried out by two people, enabling to cut needles into the measurement container already in the car during the drive to the laboratory. Thus, the time-gap between the sampling and measurements was reduced from one hour to ten minutes.

The dynamic equilibrium of radon daughters in the air depends on their generation rates from natural sources and it is specific for each region. Thus, a wire experiment was performed to reveal correctly the initial activities of radioactive elements ^{214}Bi and ^{214}Pb in the sample. An 11.5 m long wire was connected to 6 volt electricity source and it collected charged ions and particles during 6 hours. Thereafter, wire was wrapped around a suitable piece of pipe and the radioactivity spectra were registered. The temporal behaviour of the registered spectra allows to calculate the initial activities of the named elements in plants correctly. The results are presented in Table 3.2.

Thus, as a result of above-described work methodologies were created, which enable to check the results of the modelling. When the validation of results of air pollution modelling was a repetition of studies elsewhere, the validation of results of modelling of pollutant deposition due to electric field influence on dry deposition of particles-ion clusters is a novel work.

4. ASSESSMENT OF AIR QUALITY

Monitoring plays a central role in environmental management enabling to get data about real world. Assessment forms the first step in the management of air quality. Management also includes measures for reducing pollution levels, therefore the recommendations of international authorities to decision-makers are dealing mainly with management of air quality.

Monitoring is not able to quantify the patterns of air pollution both in space and time. Consequently, other methods must be used in conjunction with monitoring. The overall term for all the methods used to estimate objectively air quality is 'assessment'. The exact definition of international agreement legislated in Article 2 of framework directive (COM, 1996) states that 'assessment' is 'any method used to measure, calculate, predict or estimate the level of a pollutant in ambient air'. The following chapter gives a brief description of different assessment methods and practical experience of assessment.

4.1 Ways to assess air quality

The ways to assess air quality are well defined within international agreement laid down in COM (1996), involving objective estimation, modelling and monitoring. When the content of the last two is more-or-less understood from the term, the term 'objective estimation' requires some explanation. The process of assessment and its application for optimisation of monitoring is in more detail described by WHO (2000).

The description of air quality assessment (AQA) opportunities starts with a less financial, but most expert judgement requiring objective estimation; it continues with modelling, enabling to correlate sources and receivers, pollution levels and health effects, and cost-benefits; and ends with monitoring which is a core element in air quality management.

4.1.1 Objective estimation - the simplest and the most cost-efficient way

The meaning of the term 'objective estimation' depends on the experts performing this process. The main idea of the term is expressed by the word 'objective' - thus, we need some set of objective values on which to base the estimation. The objective estimation involves usually emission measurements and inventories since the creation of elevated pollution levels is mostly determined by the sources. Since emission inventories have become a routine procedure in the developed countries during the recent decades (see chapter 2), objective estimation usually requires only a brainstorm of a group of experts of an area.

One of the widely used tools for objective estimation is passive monitoring. There may arise a question why passive monitoring is considered under estimation of pollution levels. The answer is that exposure period of passive monitors starts with day (in polluted areas) and ends with month (remote areas). Thus, the comparison with limits of shorter time period is only possible through the estimation of ratios of short and long-term average concentrations (UBA, 1997; or determined from local measurements). In the other hand the passive monitoring is cheap and therefore widely used for screening of pollution levels and thus revealing of areas-pollutants requiring further research.

It is possible to use also other methods, such as the modelling of air masses distribution patterns (Seaman, 2000), or the interpolation of monitoring and measurements results to another location (Scaperdas, Colvile, 1999).

Some activities of routine monitoring also fall under objective estimation. For example, monitoring stations are characterised by the term 'area of representativeness', expressing how much of the surrounding pollution levels could be explained by the data of monitoring. Certainly, modelling helps to reveal the exact value of this term, but the final evaluation requires objective estimation by experts. Thus, the described way of assessment is inherently included into the planning, running and advancing processes of monitoring.

4.1.2 Modelling - the finest and the only way for risk assessment

Air pollution is a 3-dimensional phenomenon. Thus, we cannot expect that it would be possible to measure the values of pollution fields or exposure levels everywhere and for everyone. The modelling is therefore the only way to answer some basic questions arising during planning and developing economic activities, such as – 'what is the cost of air pollution?' or 'how many people are living in the areas with pollution levels exceeding the limit values?' Certainly, modelling simplifies the processes, but it still gives us an enormous amount of information impossible to obtain otherwise.

An overview of applications of modelling in the assessment of urban air quality is given by Tønnesen, Leeuw and Moussiopoulus (1998). Paper concludes that since all processes running in atmosphere have a complex and stochastic nature, often very sharp temporal and spatial gradients of pollution are produced. The selection of model for assessment depends on the time and spatial scale of issue under consideration, accuracy of input data (meteorology, obstacles, sources), requirements on reliability of modelling results, and chemical-physical processes influencing modelled species.

An important aspect of AQA is risk assessment determining priorities of further activities. The bigger is the risk to human health, the more measures are required to evaluate and minimise it. Simple risk assessment is based only on measurement results and assumes that all people are exposed to the same level of pollution as has been monitored.

4.1.3 Monitoring - the most expensive way

Monitoring is the only way to compare our predictions and estimations with the real world. For the process, a set of monitors is positioned at the selected sites to give information about pollution fields. The requirements on methods and data quality are expressed in Chapter 1, a more detailed description of monitoring has been summarised in WHO (2000) and Sivertsen (1997).

A relation in statistical physics allows to replace averaging in space with averaging in time in homogenous fields, enabling to estimate from the measured time series of pollution levels the levels of pollution around the monitoring site. It is properly correct for an 'area of representativeness (AOR)', mentioned in 4.1.1. The AOR could certainly be different for different pollutants, and it could also be different in different meteorological conditions.

In developed countries monitoring can be classified into several groups: 1) indicative monitoring - required time coverage at least 14%; 2) continuous monitoring - required time coverage at least 90%, and 3) special investigations performed for revealing of reasons of pollution, source apportionments or other mostly scientific purposes. The accuracy of measurements must in every class of monitoring correspond to the specified values (COM, 1999).

Monitoring requires large investments into instruments, people, computers, etc. Therefore, in the developed countries, monitoring activities are as much as possible replaced by less consuming methods. The next subchapter shows in which conditions monitoring is absolutely necessary.

Risk assessment has to precede monitoring activities. The usual objects of monitoring are pollutants with high health-risks and/or exposure levels. Pollutants with low health-risks and/or exposure levels are often assessed by cheaper ways described above. The discussion of priority setting has well been explained in chapter Priority Setting in Air Quality Management of WHO guidelines (2000) and will not be discussed in detail here.

4.2 Practice of air quality assessment

4.2.1 International experience

The acceleration of urbanisation and rapid increase in number of cars initiated in all countries air quality issues. Increased mortality and illness of people lead to a significant development in air pollution assessment and reduction strategies. As a result in developed countries a significant drop in sulphur dioxide and soot levels has taken place. However, increasing car fleet has initiated new air quality issues associated with nitrogen oxides and ozone (Fenger, 1999; Mayer, 1999; Colvile et al., 2001).

The standardisation and simplification of assessment process is a target of international agreement set by COM (1996), which give, in addition to limit values, also the concept of upper and lower assessment thresholds. When pollution level exceeds the upper threshold, monitoring is mandatory. In case of

exceeding the lower threshold – modelling is mandatory. Thus, pollution levels determine which kind of activities must be carried out for the assessment of air quality – see Table 4.1. Usually, some data about pollution levels have already been drawn from modelling or monitoring and thus the determining of mandatory activities and zones is easier.

It is important to note that the dimensions of zones where certain assessment activities are mandatory could be as small as one side of a busy street.

Table 4.1 Upper and lower assessment thresholds (in parenthesis the number of violations of limit allowed in any calendar year) of concentrations ($\mu g\ m^{-3}$) of pollutants set by COM (1999, 2000).

pollutant	Averaging period	Upper assessment threshold		Lower assessment threshold	
		health	vegetation	health	vegetation
Sulphur dioxide	24-hour 1 year	75 (3)	12	50 (3)	8
Nitrogen dioxide	1-hour 1 year	140 (18)	32	100 (18)	26
nitrogen oxides	1 year	24		19.5	
PM10	24-hour 1 year	30 (7) 14		20 (7) 10	
lead	1 year	0.35		0.25	
Carbon monoxide	8-hour	7 mg m^{-3}		5 mg m^{-3}	

A logical order of assessment methods in zones without any data of air quality is: first, objective estimation; second, when the levels of pollutants exceed the lower assessment threshold – modelling; and third, when the levels of pollutants exceed the higher assessment threshold – monitoring. To simplify the assessment, it has specially been stated that monitoring is mandatory in cities with population over 250, 000. Against the background of usual patterns of human activities, traffic creates pollution levels requiring monitoring of air quality in such agglomerations.

In the developed countries special schemes have been established to assess air quality an easier way, still maintaining the high quality level. The developed approaches can be summarised by the following steps: 1) objective estimation of pollution fields by total inventorying of pollution sources; thereafter, if reasonable 2) screening of pollution fields either by modelling or passive monitoring, and thereafter if violations of limit values occur - 3) pilot studies. When all steps show a high potential to exceed the limit values at some locations, a set of actions must be undertaken to obtain new information about the possible exposure of people or ecosystems and if necessary, a monitoring network should be advanced.

According to an international agreement, the number of monitoring sites and the requirements regarding the locations of monitoring stations also depend on the size of city. Thus, decision-making is made as simple as possible.

A good example of the simplification of assessment process for decision-makers is England. The United Kingdom National Atmospheric Emission Inventory (NAEI, 2002) published on the Internet enables the local authorities to assess air quality in a proper way, but easier.

The cost-efficient way to assess air quality is passive monitoring. In England such network for NO_2 was established already in 1987 (Bower et al., 1991) and is operating until today. A similar approach, including also SO_2 and hydrocarbons is used in Sweden (Svanberg et al., 1998).

The selection of model for applications is made simpler by development of special Model Documentation System (MDS, available at http://www.etcaq.rivm.nl/databases/mds.html). MDS describes in the database model parameters and its application area, thus enables potential user to choose between tens of models available on the market. Analysis of users shows that they are mostly scientists, thus the target group of administrators did not have yet recognised the usefulness of modelling (Moussiopoulos et al., 2000).

A powerful example of modelling for AQA is described by Heldstab et al. (1999). Study performed and considering both direct emissions of particles and their formation from precursors shows that 61% inhabitants of Switzerland live in areas with excessive PM10 pollution levels. Study shows also that traffic is responsible for about half of emissions of particles through emissions from tailpipe, tires, brakes, and abrasion - resuspension from road surface.

There have developed schemes to use for assessment indicative monitoring of some pollutants (VOC-s, heavy metals named in COM (1996)) requiring otherwise too many resources. In that case the even reasonable distribution within a year is very important.

Thus, international experience shows that all assessments methods are used and developed in conjunction to each other to assess air quality in a more proper way.

4.2.2 Estonian experience

The legislative framework of Estonia for the assessment of air quality is not so well established as internationally. The responsibility to perform the assessment is given to local governments with few supporting guidelines, which are copied mainly from EU directives mentioned earlier. Therefore, the level of performed assessments varies significantly and most of them consider only some pollution influencing factors.

4.2.2.1 Current practice

The usual practice of air quality assessment in Estonia relies on the data of monitoring, casual measurements, which usually did not have enough time coverage and/or quality, and the modelling of pollution caused by enterprises.

Monitoring data were taken as representing the whole city (background stations) or the highest pollution levels (traffic stations in city centre). The term 'area of representativeness' required for international EUROAIRNET stations is declared, but not used in assessing air pollution in cities. Since the above-mentioned stations operate only in the capital and at three sites in remote areas, the basis for assessing the situation in other towns and areas is weaker.

In recent years some effort has been made in using modelling for air quality assessment. In the towns of Kuressaare, Kärdla, Pärnu, Tartu, Narva, Tallinn and Viljandi pollution levels for the worst and average cases were calculated for urban planning purposes (Kaasik et al. 2001; GAIAA, 2001). The model AEROPOL mentioned in Chapter 3 was used for that purpose. Studies performed reveal that dangerous pollution levels can appear when slow wind is blowing along the busy street and thus the effect of pollution accumulation appear. Studies show also that modelling of particulate matter distribution cannot be carried on due to lack of data of pollution sources (traffic and area sources are not considered in inventories). The estimation of violation of the required assessment thresholds (see Table 4.1) is, however, difficult due to the structure of the program. Thus, the modelling results do not allow to perform the full assessment procedure.

Chapter 2 shows that the assessment of air quality by emission inventories, cannot be carried out very well in Estonia due to gaps in these inventories. Another factor that prevents objective estimation can be the lack of qualified people having interest in performing proper assessment. The above reason is supported by the fact that no risk assessment and/or source apportionment studies, enabling to evaluate the seriousness of different sources and thus to establish a more realistic background for proper monitoring structure have been performed.

There are appearing first signs of special studies such as heavy metal (HM) investigation performed in autumn-winter 1998-1999 (EERC, 1999). Unfortunately, drawbacks in methodology (non-even distribution within a year, high and variable HM levels in collection media – filter, and measuring of elements of only anthropogenic origin) did not allow to use results for source apportionment.

4.2.2.2 Scientific studies

Although the official interest to reveal reasons of monitored phenomena and predict the future trends is low, several groups still carry out scientific research. On the following pages I try to explain the main results of the studies performed, which are suitable for AQA.

Air quality measurement studies were performed at the Institute of Environmental Physics in Tartu. The main area of research has long-time been aerosol, recently also pollution modelling due to joining with meteorologists from Observatory of Tôravere.

Assessment of atmospheric aerosol concentrations shows that in a remote area the data of particles of the diameter less than micrometer represent an area

with radius of up to several hundred kilometres in wind direction and between 50 and 100 km in case of crosswise winds. Similarly, the area of representativeness of bigger particles is only some tenths of kilometres, being almost linearly proportional to particle size (Vana, Tamm, 2002).

Assessment of black carbon (soot) concentrations measured by individual aethalometers at 14 sites in the capital, other towns and villages in 1995 (Kikas et al. 1996) showed that the elevated levels are mostly associated with domestic heating. Higher concentrations of black carbon were obtained in smaller towns, most probably expressing the smaller height of stacks of boiler houses and the bigger role of residential areas with individual heating. Although concentrations did not exceed 10 $\mu g\ m^{-3}$ and, thus, the measurements of soot concentrations are not urgently required, it gives an opportunity to estimate the influence of heating activities and diesel soot on air quality at low cost. Additionally, soot concentrations enable to estimate the levels of PM10 or PM2.5 (COM, 1996; Vallius et al., 2000) or some organic compounds associated with the creation of elemental carbon in combustion processes. Thus, black carbon measurements are suitable in conjunction with passive monitors for getting information for objective estimation of air quality.

The scientific analysis of the data of air quality monitoring has been given by Kõrvits (2000) and Kimmel et al. (2002). In the named studies an analysis of the most important air pollution issues in Estonia has been made. The description of results is given in Chapter 1 and in supplementary article 4. Accordingly, the most critical air pollution issues of Estonia are dangerous levels of ozone and particulate matter. The Chapter 2 shows that for both pollutants the overview of sources is too fragmentary. Supplemented article 4 proposes that dangerous levels of particulate matter, particularly in spring, are associated with traffic. The reason may well be also salting and sanding of streets, natural emissions or other processes listed in COM (1999, 2001). The source apportionment study is the only way to end speculations, to reveal reasons of harmful level of pollution, and thus to find measures necessary for reduction of pollution.

The existing studies and experience of other countries show that the applicability of monitoring data of Estonia to the country as a whole is doubtful. For instance, for one and the same time period NO_2 levels were falling in Tallinn (see Figure 1.2), but in Tartu (Kimmel, Kaasik, 2002) an increase occured almost in the whole city.

In Estonia, air quality assessment by modelling is mainly based on the models of Russian origin, and models developed at the Observatory in Tôravere and it has been described earlier (Kaasik, Kimmel, 2000). Modelling by program AIRVIRO, developed by the Indic group and used in England, Sweden and other countries, ha intensively been performed in Tallinn, but the results have unfortunately not been published neither on local, nor on international levels.

The practice of air quality assessment by objective estimation is very rare in Estonia. It is possible to use the results of passive monitoring for the objective estimation of air quality – see Table 4.2. In Estonia passive monitoring is carried out in Tallinn by Swedish programs, in county centers of South Estonia as described by Kimmel (1998) and Kimmel, Kaasik (2002), and at the remote station in Karula (see http://www.seiremonitor.ee, left menu qhuseire). In Tallinn, NO_2, SO_2 and selected VOC-s were measured, in other towns the measurements included mainly only NO_2. The evaluation of hourly maximums and yearly averages (the time coverage was more than 14% and the measurements were evenly distributed within a year) shows that air quality requires some research in Tartu and Viljandi, since the upper assessment threshold is exceeded – see Table 4.2.

Table 4.2 Estimated concentrations of NO2 in South-Estonian county centers.

	Estimated concentration $\mu g\ m^{-3}$		
Location	Yearly average	24 hour maximum	1 hour maximum
City center of Tartu	47	150	252
City center of Viljandi	39	125	210
Crossings of main streets of Tartu	39	135	230

Sources: Kimmel (1998), Kimmel and Kaasik (2002)

The study performed for evaluating the influence of electric field on dry deposition of aerosol particles described in Chapter 3 enables to correct the assessment of deposition loads of air pollution. Tammet et al. (2002) shows that electric forces must be considered for particles with the diameter of 10-200 nm in conditions of low wind speed (< 2m s^{-1}). Thus, for the correction of the estimation of pollutant deposition on plants we require data of: 1) repeatability of wind speed at the height of the target plant, 2) the ratio of dry/wet deposition and the chemical composition of different size fractions of aerosols at the site, 3) the deposition loads of pollutants in an open area. Preliminary assessment shows that 1) low wind is most common in South and Central Estonia (Kull, 1996), 2) no data exists about the chemical composition of different aerosol size fractions, 3) the analysis of critical loads of acidifying components by Oja, Kull (1998) and Oja (2000) shows that the critical loads are exceeded in South-East Estonia. Thus, concluding from the findings of the preliminary assessment, the pollution loads are most critical to ecosystem in Central and South Estonia. Consequently, the further research is reasonable.

Another opportunity used for the assessment of pollution levels is wind field modelling. The repeatability of wind speed and direction in Tartu was calculated by WASP program (Mortensen et al. 1993) as described by Kull (1995) and Karu (1996). The modelling of wind field supported by a round the clock measurement campaign of characteristics of wind field shows that in

Tartu, the almost stable atmosphere with no movement of air masses is often formed in the valley of the River Suur-Emajõgi flowing through the centre of city. Thus, a good opportunity for pollution accumulation is created.

4.2.2.3 Simple scheme developed for a medium-sized town

The unsatisfactory overview of air quality in Tartu triggers the development of a simple scheme for the assessment of air quality in all the territory of city. The detailed description of the scheme developed is given in the supplementary article III. The scheme consists of: 1) wind field modelling for prediction of zones with favourable conditions for pollution accumulation, 2) passive monitoring of pollution fields prevalently in revealed zones, 3) updated overview of air pollution sources (mainly domestic heating and traffic counts), 4) modelling of pollution distribution by simple Gaussian-plume model AEROPOL.

The scheme enables to measure long-term averages of pollutants at the same time in number of places, and simulate yearly averages and maximum concentrations of pollutants. Thus, a methodology to evaluate the influence of economic projects on air quality, and predict changes in future was created.

The first good result of scheme was a relocation of monitoring station closer to area with the highest pollution levels. However, since the relocation occurred in 1998, and changes in traffic schemes and emissions patterns were happened also later (see supplemented article III), the current location may need already some corrections.

4.2.2.4 Summary of Estonian experience

Examples given in last pages could be a little confusing for readers. Therefore, the following Table 4.3 and scheme explains the overall situation of air quality assessment in Estonia in a more readable form.

Table 4.3 Qualitative overview of aspects of classical air pollution in Estonia (‘±’ - mostly OK, requires some improvement; ‘-+’ - requires improvement, some part OK; ‘-‘ requires significant improvement)

		Completeness of information available	Estimation of risks associated
Emissions of	particles	-+	-
	gases	±	±
Distribution of	particles	-+	-+
	gases	±	±
Pollution levels of	particles	-	-+
	gases	-+	±

Table summarises that we do not have enough information of pollution in Estonia. The most critical seems to be lack of information about particulate

matter, which is according chapter 1 one of the most dangerous pollutant in Estonia.

Scheme 4.1 Situation of air quality assessment in Estonia.

Information about pollutants	People involved
Monitored in some cities. Other places - casual measurements. Modelling activities in towns rare. Overview of emission sources requires improvement (real emissions rarely measured, uncertainties of official values unknown). List of pollutants monitored does not correspond to the associated risk and/or the requirements of international networks.	Experienced staff of monitoring. Too few modellers. Weak links between scientists and officials. Responsibilities of local state authorities not clearly regulated- legislated.

4.3 Selection of rational approach for air quality assessment in Estonia

The use of term 'rational' in the present Thesis requires some clarification. The target of rational approach can be cost-efficiency (i.e. more information with the same cost), comfort of people (i.e. less changes) or institutions (i.e. the larger the turnover the better) involved, etc. In the current paper the target of rational approach (optimisation in practical meaning) is cost-efficiency.

Table.4.4 Qualitative overview of AQA methods in Estonia

('+' – OK; '±' - mostly OK, requires some improvement; '-+' - requires improvement, some part OK; '-' - requires significant improvement)

	Estimation		Modelling	Monitoring	
	Screening	Emissions inventory		continuous	indicative
Stage of development	-	-+	+-	+	-
Financing	-	+-	-+	+	-
Recent development	-	+	+-	+	_+

The most expensive general method of air quality assessment is monitoring. Thus, the main subject of the subchapter is the discussion of measures balancing AQA methods.

Table 4.4 shows why the special subchapter is created and shows where the investment is most reasonable.

Optimisation of air quality assessment is a question of political concern. However, scientific methods enable to evaluate the current situation and the future trends more precisely.

4.3.1 Measures required for sound optimisation of air quality assessment

Since spatial coverage of measurements in Estonia is too scarce (see Figure 1.1), passive monitoring in conjunction with the measurements of black carbon as described by Kikas et al. (1996) can provide a picture of pollution levels. In such a way we can get also spatially relevant information enabling to estimate PM10 and PM2.5 levels.

Analysis of direction of air masses carrying pollution shows that transboundary fluxes are mostly influencing South and West Estonia. Since in South does not have any advanced monitoring station and AOR of remote stations does not exceed usually 100 km, it may be reasonable to put one remote station in Karula. The proposed location was approved by coordinators of EMEP – NILU already in 1996, and supported also by AQMEU (2001), which states that in South and Central Estonia 5 additional air quality stations must be installed in remote areas.

One important factor in the optimisation of AQA is the qualified analysis of all collected data of monitoring, modelling and objective estimation. A significant improvement in analysis of data (arguing of statements, back-trajectories for pollution origin estimation, etc.) during recent years shows that the quality of analysis is coming better. There still exist some strange claims in reports, such as ‘there exist no sources of ozone in urban area’ (EERC, 2002), but they are rare. Ozone is formed in photochemical reactions from nitrogen oxides and hydrocarbons – both pollutants can be found in abundance in urban air. The reason for lower levels of ozone in urban areas is determined by existence of many sinks, not available in remote areas or less available in suburbs, which destroy the formed ozone quickly.

The analysis of air pollution sources in Chapter 2 showed that many important source classes, especially for the most harmful pollutants such as particulate matter and volatile organic compounds, were until recent years not considered in inventories. Analysis of work performed in the area shows that together with inventorying of enterprises, estimation of pollution sources producing much more pollution than enterprises can significantly improve our overview of emissions. Fortunately, in the last years the staff of inventorying

has increased and thus many important pollution sources such as domestic heating and biogenic emissions of hydrocarbons are already considered (see Chapter 2). We hope that such improvement occur also with emissions of particles from surface due to wind and traffic.

It is possible to use also mathematical modelling of pollution fields for the optimisation of air quality monitoring. When the difference between the estimated and measured values of the simulated parameter gets too large, it is reasonable to start monitoring.

An example of a theoretical approach has been given by Trujillo-Ventura and Ellis (1991), showing how multi-objective air pollution monitoring can be carried out with the help of mathematical approximations and trade-offs between various parameters. However, theoretical approximation requires very clear definitions of the initial situation, restrictions and targets, which can be difficult to obtain.

The easier and more practical way for smaller countries is to use the experience of the developed countries.

4.3.2 Possible use of methods developed for optimisation of air quality assessment

The methods developed in the present thesis can be used for the optimisation of air quality assessment. To simplify their application in everyday activities the thesis gives a description of their potential use. The introduction of the methods is followed by the explanation of their potential use, which is not associated with their importance.

The advanced analysis of monitoring results enables to evaluate the completeness and necessity of the lists of pollutants. When the analysis shows a high potential of some pollutants (like ozone and particulate matter in Estonia) to exceed the limit values it is reasonable to arrange a special investigation to reveal the extent of the problem. When a pollutant's levels are much lower than the limits, some other techniques could be used for its assessment, such as modelling, temporary monitoring or estimation.

The analysis of the data of emissions enables to evaluate the magnitude of risks associated with air pollution. The revealing of target areas, where efforts in reducing the amounts of pollution and/or increasing the accuracy of the values of the emissions are the most cost-efficient, is an important achievement. The analysis has shown how sources not included in official statistics (such as natural emissions of organic compounds and particulate matter; domestic heating) can significantly contribute to the elevated pollution levels, and must be considered and targeted also in the emission abatement and reduction schemes.

The method of calculating the influence of the electric field on dry deposition of particles on the top branches of trees enables to evaluate the

areas, where the risks to enhanced deposition of pollutants and thus the risks to damage to ecosystems is higher. Such information in conjunction with the actual deposition loads monitored in open areas can be used for locating the sites where the monitoring of deposition is most reasonable and necessary.

The simple scheme developed for the assessment of air quality in medium-sized towns can simplify the assessment of air quality in most towns of Estonia and can thus, significantly enhance our understanding of the seriousness and extent of air pollution issues at a low-cost.

The analysis of scientific research performed in Estonia has shown that many of these activities can help to organise the assessment of air quality in a more proper way.

Summarising the results of the chapter we can say that balanced development of air quality assessment methods seems to be the most rational approach for developing countries helping to manage air quality in accessible way.

SUMMARY

The aim of the present thesis is to test and improve different methods of air pollution analysis with respect to their application in environmental management in a small and rapidly developing country. The analysis should provide ideas for the optimisation of air quality assessment under strongly limited resources.

The dynamic system of Air Quality Monitoring of Estonia is described and analysed in Chapter 1. The following conclusions have been drawn: 1) the continuous AQM system of Estonia is spatially too sparse, it has been started only in 1994 and it does not allow to assess air quality in the whole territory of Estonia; 2) intermittent monitoring data did not correspond to the quality criteria set by (Lalas and Saeger, 1998) and they are not sufficient for proper assessment of air quality in Estonia; 3) during the last decade the development of monitoring has been hindered by the shortage of financial resources and it has rather been controlled by occasional support from international sources.

Chapter 2 describes the categorisation of air pollution sources and the calculation of emissions from the categorised sources according to the internationally established CORINAIR scheme. Scientific research (Kaasik et al., 2001) has shown that the official data cover less than a half of all anthropogenic emissions on the local level. One of the discrepancies between the Estonian practice and the CORINAIR scheme is the calculation of emissions from mobile sources. A simplified CORINAIR scheme developed by the author for the calculation of emissions from mobile sources is a response to that demand (Kimmel, 1998). The simple scheme relies on fuel consumption data and the splitting of vehicles into classes. The scheme gives satisfactory results in comparison with the later full CORINAIR calculations (COWI, 2000). Thus, the scheme can be used for the correction of the previous emissions data. Additionally it is shown that natural emission of particles and volatile organic compounds are significantly responsible for the dangerous pollution levels of particulate matter and ozone. An estimation of natural emissions performed by the author shows that these emissions are comparable to the anthropogenic sources of pollution in Estonia. Thus, natural emissions of pollutants must be seriously considered in the assessment of air pollution in Estonia.

Air pollution models and the validation of modelling results are discussed in Chapter 3. The chapter consists of two sections: 1) the spatial validation of the predictions of pollution fields and 2) the validation of the predictions of electric field influence on dry deposition of fine particles. In the first part at the chapter a system of passive monitoring in Estonia is proposed and tested using the air pollution dispersion model AEROPOL. A fair agreement was found both in urban and background areas. In the second part a model of dry deposition of airborne particles is developed including the effect of electrostatic precipitation

of pollutants on the sharp needles that concentrate the atmospheric electric field. Radon daughters were selected as the markers in the validation experiments. The scheme consists of the selection of isotopes, the quick preparation of plant samples, the measuring of gamma spectra and the analysis of obtained data. The concentrations of radioisotopes on the needles of the top branches appeared several times higher than within the tree, confirming the theoretical predictions described in the supplemented article I.

Chapter 4 describes the strategies for the air quality assessment (AQA) and their application in practice. The author's contribution to the area is associated with the analysis of monitoring data (see Chapter 1), the developing of methods for the calculation of emissions (see Chapter 2) and for the validation of the results of the air pollution model (see Chapter 3), and the estimation of pollution levels (see PM10 estimation in Chapter 1 and Chapter 4). The simple scheme developed for the assessment of air quality and applied in Tartu is a cost-effective opportunity for proper assessment of air quality in the whole territory of medium-sized towns with limited resources (Kimmel, Kaasik, 2002). The analysis of the findings of the previous chapters shows that despite the monitored dangerous levels of ozone and particulate matter in Estonia (similarly to Europe, COM, 2001), no studies have been performed to reveal the area of concern and the reasons of the problem. The last subchapter describes the opportunities for optimisation of AQA in Estonia and finds that balanced development of different AQA methods is the most reasonable way. The analysis of the factors limiting the development of a sound, cost-effective and efficient AQA system in Estonia shows that there is a lack of objective information about emissions and pollution levels, and of interest of the authorities. The last page describes the possible ways of use of the methods developed for the optimisation of AQA.

ÕHU KVALITEEDI HINDAMISMEETODITE ANALÜÜS. MEETODITE RAKENDUSED EESTIS (KOKKUVÕTE)

Käesolev töö esitab kokkuvõtte autori poolt viimase 8 aasta jooksul teostatud õhusaaste uuringutest.

Õhusaaste on pideva linnastumise tingimustes muutunud üheks enim inimesi mõjutavaks keskkonna aspektiks. Seetõttu on ka mõistetav teemat käsitlevate rahvusvaheliste kokkulepete (COM, 1996, 1999,2000), uuringute (vt. ülevaaadet WHO, 1987, 2000) kasvav hulk. Kõigi nende dokumentide aluseks on teadusuuringud ning neid täiendatakse vastavalt uute uuringute tulemustele.

Samas on nii riikide kui linnade tasemel piirkonna arengu otsustajate töö muutunud järjest keerulisemaks. Kasvav informatsiooni hulk saastetasemeist ja saastajatest, üha keerulisemaks muutuvad seosed inimeste tervise ja õhku paisatud heitkoguste vahel ei võimalda enam adekvaatselt tajuda otsustest tekkivaid muutusi ning planeerida elukeskkonda meid kõiki rahuldaval kombel. Seega on õigete otsuste tegemisel eluliselt vajalikud uuringud kohalikul tasandil olulistest faktoritest ja nende arvestamise võimalustest.

Õhu kvaliteedi – vastavuse kehtestatud normidele- hindamiseks on kasutusel 3 üldisemat metoodikat:

1) õhusaaste seire kohtades, kus saastetasemed mõjutavad oluliselt inimeste tervist, ökosüsteeme, materjale;

2) õhusaaste allikate ja sadenemisvoogude seire;

3) õhusaaste tekke, leviku ja sadenemise modellerimine ning saastetasemete hindamine emissioonide ja saasteainete vaheliste seoste alusel.

Nende üldisemate metoodikate täitmine kohalikule tasandile olulise sisuga on juba iga riigi valik ja kohalike keskkonnauurijate tegevus.

Õhusaaste seire alused, Eesti seiresüsteemi kirjeldus ja seiretulemuste analüüs on esitatud dissertatsiooni 1. osas. Analüüs näitab, et ohtlikuimad saasteained on tahked osakesed ja osoon. Samas ei ole seiretulemused kas ruumilise kaetuse või kvaliteedist tingituna piisavalt esinduslikud õhu saastetasemete hindamiseks kogu Eestis.

Õhusaaste heitkoguste arvutamine ja ülevaade õhusaaste allikatest Eestis on 2. osa teemad. Saasteallikate analüüs baseerub rahvusvahelisel saateallikatest ülevaate saamise standardil CORINAIR. Teises osas on kirjeldatud autori poolt loodud liikuvvahendeist heitkoguste arvutamise metoodika liikuvvahendite läbisõidu andmete puudumisel (Kimmel, 1998). Põletatud kütuste kogustest lähtuv metoodika jaotab liikuvvahendid klassidesse ja annab võrreldes täiuslikuma CORINAIR standardiga (COWI, 2000) piisavalt täpse tulemuse. Seega võib metoodikat edukalt kasutada varasemate liikuvahendeist heitmete arvutustulemuste, mis on saadud Vene metoodikal ilma liikuvvahendeid klassidesse jaotamata, korrigeerimiseks.Samas osas leiab kajastamist samuti

looduslike allikate kui oluliste tahkete osakeste ja osooni tekkimisel osalevate lenduvate orgaaniliste ühendite heitmete arvutus. Autori poolt teostatud looduslike allikate õhuheitmete arvutus näitab et need on vähemalt võrreldavad inimtekkeliste heitkogustega ja seega vajavad kindlasti kajastamist õhusaaste hindamisel Eestis.

Õhusaaste modelleerimine ja mudelite tulemuste kontrolli metoodikate väljatöötamine on käsitletud 3. osas. Osa jaguneb saasteväljade modelleerimistulemuste ja elektrivälja mõju ioonide-laetud aerosooliosakeste kuivsadenemisele kontrolliks. Saasteväljade mõõtmiseks on pakutud passiivseid kogujaid ja kasutatud neid AEROPOL abil saadud õhusaaste modelleerimistulemuste kontrolliks. Tulemused olid heas kooskõlas nii linna- kui foonialadel mõõdetud NO_2 ja SO_2 kontsentratsioonidega. Aerosooliosakeste kuivsadenemise uurimiseks arendati mudel, mis kajastab elektrostaatilist saasteainete sadenemist tervatele okastele kui atmosfääri elektrivälja suure gradiendiga aladele. Mudeli tulemuste kontrolliks valiti radooni tütarelemendid nende füüsikaliste omaduste ja hea mõõdetavuse tõttu. Arendatud metoodika koosnes isotoopide valikust, taimestiku proovide kiirest ettevalmistusest, proovide gammaspektrite mõõtmisest ja mõõtetulemuste analüüsist. Uuringute tulemusena selgus, et raadioisotoopide kontsentratsioonid võrast väljaulatuvatel okstel on mitmeid kordi suuremad kui võras, mis kinnitab juurdelisatud artiklis I detailsemalt esitatud mudeli ennustusi.

Õhusaaste hindamise strateegiaid ja nende rakendamist praktikas kirjeldab 4. osa. Autori töö antud valdkonnas hõlmab seiretulemuste analüüsi (täpsemalt 1. osas), õhuheitmete arvutusmetoodikate arendamist (täpsemalt 2. osas), modelleerimise tulemuste kontrolli metoodikate arendamist (täpsemalt 3. osas) ja saastetasemete hindamist (PM10 saastetasemete hinnang 1. osas ja 4. osa). Eelmiste osade tulemuste analüüs näitab, et hoolimata tahkete osakeste ja osooni ohtlikest saastetasemeist (analoogselt Euroopaga, COM, 2001) ei ole veel soovitud korraldada uuringuid, mis selgitaksid antud probleemi ulatust ja põhjusi. Artiklis (Kimmel, Kaasik, 2002) täpsemalt kirjeldatud lihtne õhusaaste hindamise skeem on efektiivne õhusaaste hindamise võimalus keskmise suurusega linnades piiratud ressursside korral. Neljas osa kirjeldab ka kuidas on teadusuuringute tulemused rakendatavad õhu kvaliteedi hindamiseks.

Õhu kvalteedi hindamise optimeerimine on 5. osa teema. Efektiivse, põhjendatud ja majanduslikult otstarbeka hindamissüsteemi arengut Eestis limiteerivate tegurite analüüs näitab, et peamisteks kitsaskohtadeks on objektiivse informatsiooni nappus heitkogustest ja saastetasemeist ning otsustajate huvipuudus. Huvipuuduse heaks väljenduseks on hindamissüsteemi efektiivsemaks muutmist kajastavate uuringute puudus. Viimane alaosa käsitleb võimalusi autori poolt arendatud meetodite kasutamiseks õhu kvaliteedi hindamise optimeerimiseks.

Peamised töö tulemused on:

1) Eesti õhuseire jaamade ruumiline asetus ja seiratavate parameetrite valik vajab täiendamist,

2) Eesti atmosfääriõhu kvaliteet 1994-2001 vastab valdavalt nii Eesti kui Euroopa standardeile. Suurim normide ületaja on osoon taustaaladel,
3) Autori poolt loodud liikuvvahendite emissioonide arvutamise lihtsustatud CORINAIR skeemi saab kasutada varasemate emissioonitulemuste, mis saadud ilma liiklusvahendeid klassidesse jagamata, korrigeerimiseks.
4) Atmosfääri elektriväli aktiveerib peenete aerosooliosakeste sadenemist puude ladvaokstele. Efekti kvantitatiivseks kontrolliks arendati gamma-spektroskoopiline radooni tütarelementide, mis sadestatud peenete aerosooliosakeste või klasterioonidega lehtedele-okastele, määramismetoodika. Elektrilise sadenemise toime ilmnes eriti selgelt kõrgepingeliinide aluste puude ladvaokstel.
5) Lihtsad vahendid nagu seire passiivsete kogujatega, õhusaaste hajumise modelleerimine Gaussi-jaotusega, saasteallikate täiendav inventuur ja liiklustiheduste loendused moodustavad sobiva aluse õhu kvaliteedi hindamiseks keskmise suurusega linnades nagu Tartu.
6) Õhu kvaliteedi hindamise optimeerimine nõuab kõikide saaste tekkimise-leviku-sadenemise aspektide arvestamist. Neid aspekte saab arvesse võtta saasteallikate inventuuride, meteo- ja saasteväljade modelleerimise, seire ning teadusuuringute andmete analüüsiga.

Juurdelisatud publikatsioonid käsitlevad uuringu aspekte detailsemalt: elektrivälja mõju peenete aerosooliosakeste kuivsadenemisele (Tammet, Kimmel, Israelsson, 2001), õhusaaste modelleerimine ja tulemuste kontroll linnatingimustes (Kaasik, Kimmel, Kaasik, 2001), Eesti õhusaaste 1994-1999 tulemuste analüüs (Kimmel, Tammet, Truuts, 2002), ja õhu kvaliteedi hindamine lihtsate vahenditega (Kimmel, Kaasik, 2002).

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