



## **OSAMAT – post-project environmental monitoring in 2014 and 2015**

### **Final report**



2015  
National Institute of Chemical Physics and Biophysics  
Estonia

1. Foreword.....	3
2. Introduction.....	3
3. Materials and methods .....	4
3.1. Sampling .....	4
3.2. Meteorological data.....	6
3.3. Analysis of soil samples .....	7
3.4. Analysis of surface water samples .....	8
3.5. Legal limits .....	9
3.6. Previous monitoring .....	11
4. Results .....	14
3.7. Soil.....	15
3.8. Surface water .....	17
5. Summary of the results and trends.....	23
5.2 Soil.....	23
5.2 Surface water .....	25
5.2.1 Indicative parameters .....	25
5.2.3 Trace elements.....	33
5.3 Groundwater .....	42
6. Data analysis .....	42
7. Conclusions.....	45
8. References.....	47
Appendix 1	Accreditations
Appendix 2	Sampling reports and results of analysis
Appendix 3	Terms of Reference of the Contract

## 1. Foreword

A contract between Eesti Energia Ltd and the National Institute of Chemical Physics and Biophysics was signed in 10.06.2014 to conduct a follow-up monitoring of pilot road sections of OSAMAT-program in 2014-2015.

Purpose of the contract was to evaluate and analyze environmental status of Narva-Mustajõe and Simuna-Vaiatu pilot road sections constructed by using partly oil shale ash (OSA). Second objective of the work is to verify possible environmental effects of utilizing the OSA in larger scale.

The environmental follow-up program consisted of analyzing of surface water and soil samples in order to validate the release of potentially toxic trace elements and selected anions from pilot road structures to the environment. Based on the results of follow-up monitoring it is possible give scientific data for assessing the environmental impacts and evaluation of potential risks associated with construction of roads with OSA material.

The final report presents and discusses the results of current environmental monitoring of the pilot sections during 2014-2015 and draws conclusions based on the results.

## 2. Introduction

The preferred management option for ash flow from industrial combustion of solid fuels is utilization rather than landfilling. The production divisions of the Eesti Energia Narva power plants utilize Estonian oil shale (kukersite) to produce a total of about 9 TWh of electricity each year. At present, two different oil shale combustion processes are in use: pulverized firing (PF) and circulating fluidized-bed (CFB) combustion technologies. Due to high mineral content of kukersite (45-47 mass%) about six million tons of oil shale ash (OSA) is produced annually [1], which is mainly deposited in ash fields near to power plants. The reasons for low-level utilization of OSA in Estonia include inconsistency in the quality of OSA, high cost of transportation and partly by the fact that according to Estonian legislation OSA is considered as a hazardous waste [2] due to highly alkaline reaction in contact with water. At the moment the OSA deposits are huge and contain high volumes of important raw materials for civil engineering purposes [3].

The strategy of utilizing combustion ash in the entire world is in constant change. On one hand, it is advantageous to make commercial use of the by-products of energy production. On the other hand, the large-scale usage of very complex wastes like OSA raises questions about environmental impact of such activities. Thus, it is essential to monitor the environmental aspects of the process where OSA is brought back into the nature, including the fate of toxic elements.

In general, for risk assessments connected with utilizing of ash in road construction, it is important to know which compounds could be released into the environment. The leaching of soluble constituents upon contact with water is regarded as a main mechanism of release, which results in a potential risk to the environment. OSA contains major matrix elements such as Ca, Si, Al, and Fe together with significant amount of minor elements, *e.g.* Mg, K, P, S and negligible amounts of trace elements [4].

Recent study [5] leave us believe that the release of potentially harmful compounds, such as heavy metals from OSA is relatively low and that the OSA is safe for use civil engineering. Still, the release of toxic heavy metals is strongly pH dependent [6] at conditions prevailing in soils and the release of toxic substances could become an environmental concern. Beneficial use of the OSA in road construction requires better knowledge of leaching of soil-ash systems in order to evaluate mobilization potential of metals during soil-ash water interaction. Therefore it is essential to monitor content of hazardous elements in water and soil in areas of large-scale utilization of OSA whenever possible.

The current environmental quality criteria for OSA to be utilized in bulk quantities are rather strict in Estonia. The follow-up monitoring program evaluates the impact to surrounding environment from a road sections constructed partly with OSA material from Narva Power Plants by comparing the content of selected parameters to natural background and legal limits.

### 3. Materials and methods

The pilot sections were constructed on two roads in Estonia in Narva-Mustajõe and Simuna-Vaiatu road (see osamat.ee). In the road sections three types of OSA from Baltic Power Plant were used

- Cyclone ash from oil shale pulverized firing boiler (Cyclone PF) 5% mixed with cement 5% + aggregates (oil shale mining waste aggregate and milled asphalt concrete)
- Ash from the electrostatic precipitator of oil shale pulverized firing boiler (EF PF) 6% mixed with cement 3% + aggregates (oil shale mining waste aggregate and milled asphalt concrete)
- Ash from the 1<sup>st</sup> field of electrostatic precipitators of oil shale circulated fluidized bed boiler (EF CFB) 9% mixed only with aggregate (oil shale mining waste aggregate and milled asphalt concrete)
- Complex stabilization (traditional method) with composite cement CEM II / B-M (T-L) 42.5R.

The recipes and stabilization methods used in road sections are described in progress report no. 4 of Osamat project (2013) and are also presented in Figure 1 for Narva-Mustajõe pilot section.

#### 3.1. Sampling

Follow-up monitoring campaigns were carried out in both pilot sections in Narva-Mustajõe and Simuna-Vaiatu road in 2014 and 2015.

Two soil-sampling episodes were carried out by the specialists from Latvian Environment, Geology and Meteorology Centre by the accredited soil sampling method ISO 10381 (Accreditation see Appendix 1). Soil sampling campaign was conducted in 22.07.2014 and 22.07. 2015 (Figure 1). Soil samples were taken from the depth of 0.2-0.4 m and 0.05-0.2 m from the surface in 2014 and 2015 campaign, respectively. Exact co-ordinates of sampling points were following:

NM-1	59° 19' 45.23" N and 27° 56' 23.49" E
NM-2	59° 19' 56.62" N and 27° 57' 11.18" E
NM-3	59° 19' 59.55" N and 27° 57' 23.24" E
SV-2	59° 01' 19.5" N and 26° 25' 21.7" E
SV-3	59° 01' 09.9" N and 26° 25' 30.8" E

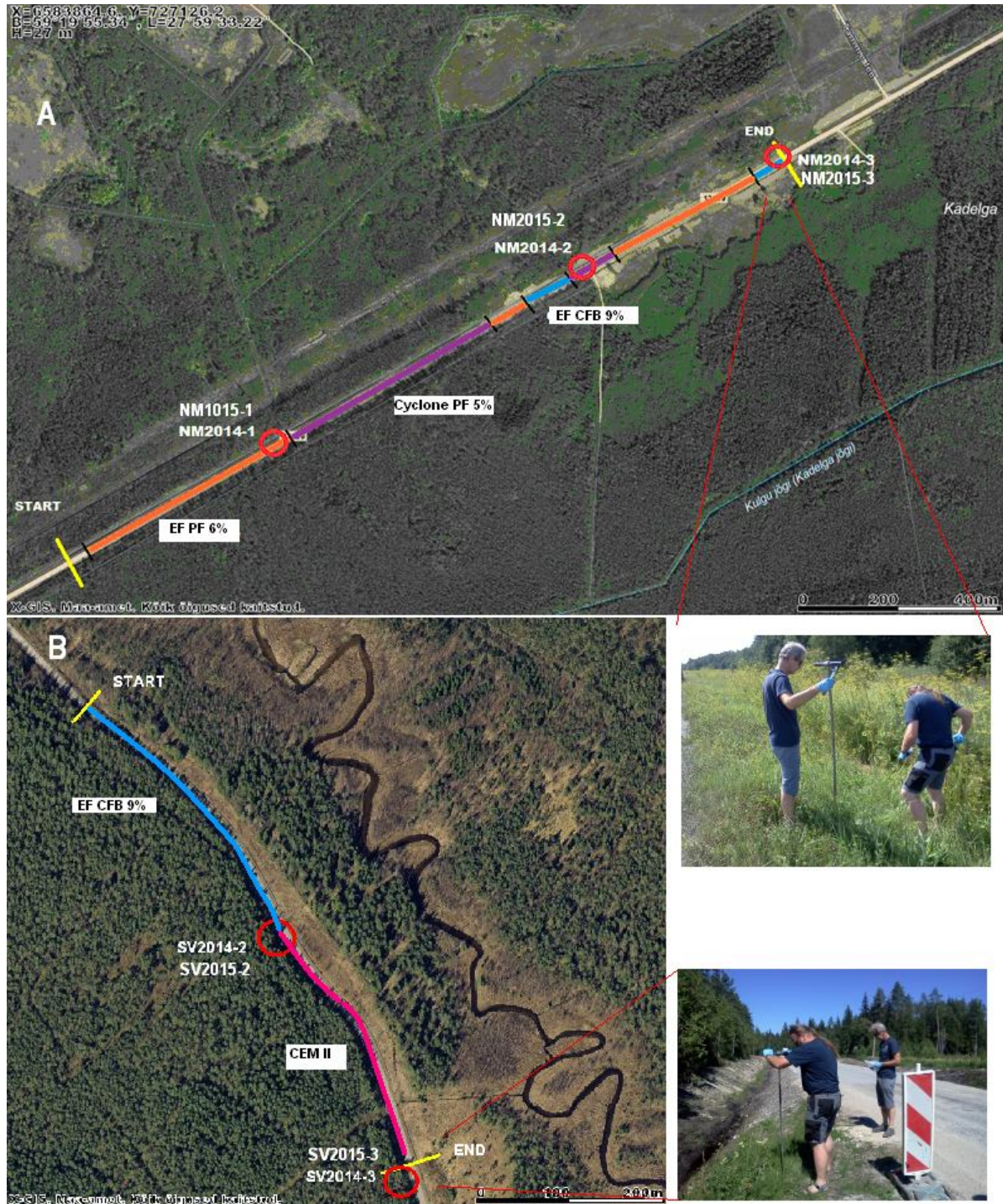


Figure 1 Locations of soil sampling points in Narva-Mustajõe (A) and Simuna-Vaiatu pilot sections.

Surface water samples were taken by the specialists from Tallinna Vesi Ltd by the accredited method EN 5667 (Attestations No. 948/11 and 1260/15, Accreditations see Appendix 1). Sampling campaigns were conducted in following dates 27.05, 30.07 and 22.10 in 2014 and 03.02, 14.04, 21.07 and 20.10 in 2015. In addition one surface water sampling episode in Narva Mustajõe pilot section was conducted in 24.11.2015 using accredited method EN 5667 by the specialist of Estonian, Latvian and Lithuanian Environment Ltd. (Attestation No. 1110/13). Location of surface water sampling sites in Narva-Mustajõe and Simuna-Vaiatu pilot sections in current monitoring and also earlier sampling locations are presented in Figure 2. During monitoring period it was not possible to take surface water samples in the end of pilot section in Narva-Mustajõe road as stated in the terms of reference due to empty ditch. It was decided to take the sample in the end of the waterfront on the other side of the road (ca 20 m from the culvert). Sampling reports are presented in Appendix 2. Exact coordinates of sampling points were following:

NM-1	59° 19' 45.23" N and 27° 56' 23.49" E
NM-2	59° 19' 56.62" N and 27° 57' 11.18" E
NM-3	59° 19' 59.55" N and 27° 57' 23.24" E
SV-1	59° 1' 27.14" N and 26° 25' 08.32" E
SV-2	59° 1' 19.98" N and 26° 25' 22.21" E
SV-3	59° 1' 14.51" N and 26° 25' 28.42" E

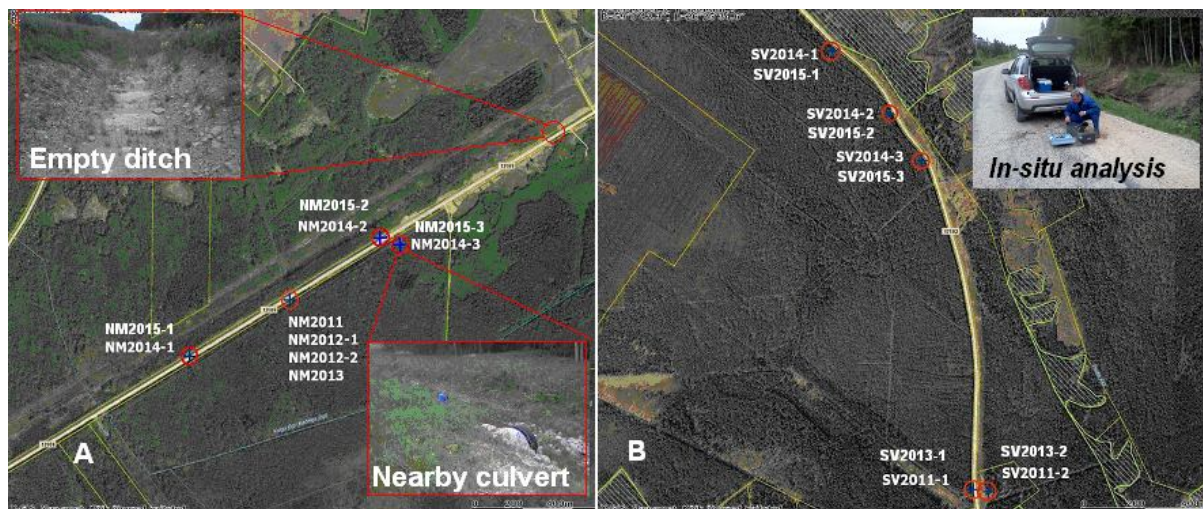


Figure 2 Locations of surface water sampling points in Narva-Mustajõe (A) and Simuna-Vaiatu (B) pilot sections in current monitoring program and in previous ones.

### 3.2. Meteorological data

Data for monthly precipitation and average temperature during the surface water-sampling period of the follow-up monitoring program (May 2014–October 2015) is obtained from the following closest meteorological stations to the pilot sections (Estonian Weather Service web page: [www.ilmateenistus.ee](http://www.ilmateenistus.ee)):

For Narva-Mustajõe pilot section - Narva station N 59°23'22" E 28°06'33", distance ca 10 km,

For Simuna-Vaiatu pilot section – Väike-Maarja N 59°08'29'' E 26°13'51'', distance ca 10 km.

The stations are equipped with automatic precipitation sensors and the observation data is presented graphically in Figure 3.

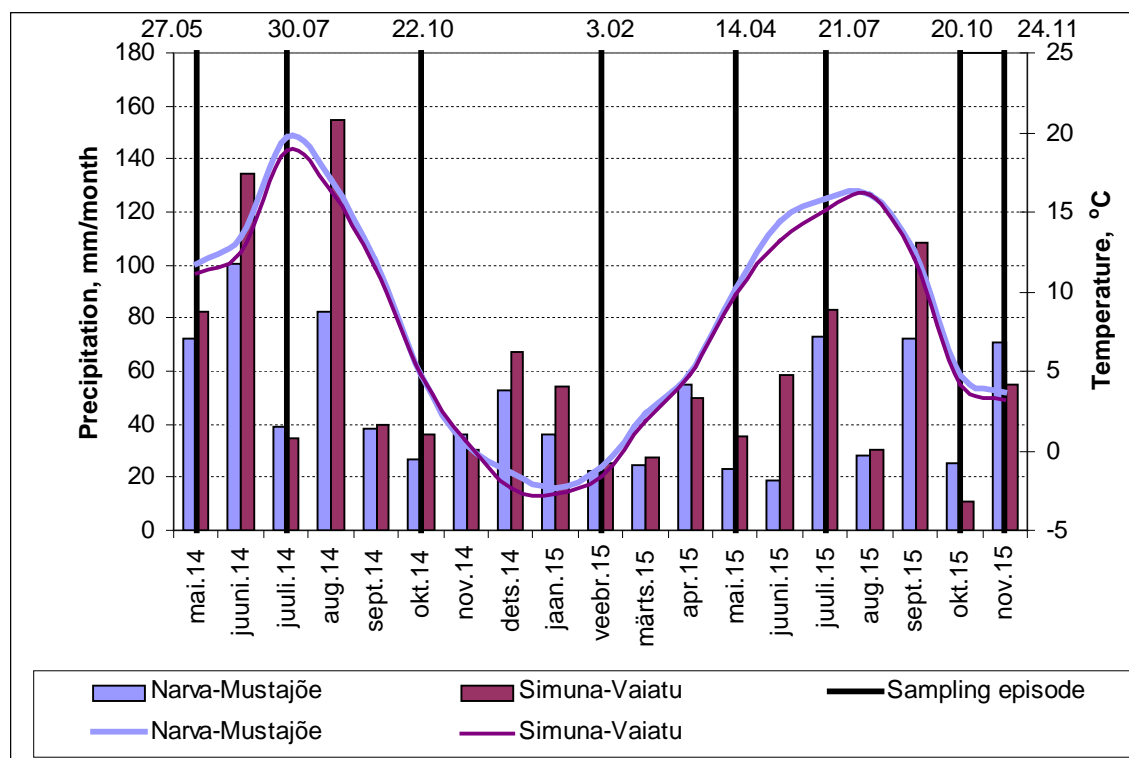


Figure 3. Monthly precipitation and average temperature in pilot road section areas as well as surface water sampling dates

Data from: (<http://www.ilmateenistus.ee/ilm/ilmavaatlused/vaatlusandmed/oopaevaandmed>).

As can be seen from Figure 3 almost all surface water samples were taken in a periods of relatively low precipitation. In that regard, of course, hindsight is a wonderful thing.

### 3.3. Analysis of soil samples

Soil samples were analyzed in the laboratory of GBA Gesellschaft für Bioanalytik mbH (Pinneberg, Germany) by accredited methods (see Table 1 and Appendix 1). Dry weight was determined by ISO 11465 with limit of quantification 0.40 wt%. Soil samples were digested according to EN 13657 and the determination of trace elements was carried out in diluted acid extracts of the samples according to following accredited methods (see Appendix 1):

Table 1 Selected trace elements analyzed in soil samples in the laboratory of GBA.

Element	Unit	Limit of quantification	Uncertainty (%)	Method
Mercury, Hg	mg/kg <sub>DW</sub>	0.10	3.00	EN ISO 16171
Cadmium, Cd	mg/kg <sub>DW</sub>	0.10	7.60	EN ISO 16171
Lead, Pb	mg/kg <sub>DW</sub>	1.0	7.20	EN ISO 16171
Nickel, Ni	mg/kg <sub>DW</sub>	1.0	7.60	EN ISO 16171

Element	Unit	Limit of quantification	Uncertainty (%)	Method
Antimony, Sb	mg/kg <sub>DW</sub>	1.0	5.00	EN ISO 16171
Arsenic, As	mg/kg <sub>DW</sub>	1.0	7.20	EN ISO 16171
Barium, Ba	mg/kg <sub>DW</sub>	1.0	5.80	EN ISO 16171
Chromium, Cr total	mg/kg <sub>DW</sub>	1.0	8.70	EN ISO 16171
Copper, Cu	mg/kg <sub>DW</sub>	1.0	nd	EN ISO 16171
Molybdenum, Mo	mg/kg <sub>DW</sub>	1.0	4.40	EN ISO 16171
Vanadium, V	mg/kg <sub>DW</sub>	1.0	nd	EN ISO 16171
Zinc, Zn	mg/kg <sub>DW</sub>	1.0	2.60	EN ISO 16171
Selenium, Se	mg/kg <sub>DW</sub>	2.0	8.60	EN ISO 16171

nd – no data.

### 3.4. Analysis of surface water samples

Following parameters were measured *in-situ* by accredited methods during the sampling episodes (see Table 2, Appendix 1):

Table 2 Indicative parameters of surface water samples determined *in situ*.

Parameter	Unit	Uncertainty, $k=2$	Method
pH		0.2	EN ISO 10523
Electric conductivity, EC	$\mu\text{S}/\text{cm}$	3%	EN 27888
Temperature	$^{\circ}\text{C}$	0.2	VL-ANP-34

In sampling episode in 24.11.2015 YSI Professional Plus Multi-Parameter Water Quality Meter was used (pH accuracy  $\pm 0.2$  units, EC accuracy  $\pm 0.5\%$ , temp. accuracy  $\pm 0.2$   $^{\circ}\text{C}$ ).

Following anions were analyzed in the laboratory on Tallinna Vesi Ltd by accredited methods (see Table 3, Appendix 1)

Table 3 Anions quantified in surface water samples in the laboratory of Tallinna Vesi Ltd

Anion	Unit	Limit of quantification	Uncertainty, $k=2$	Method
Chloride	mg/L	0.05	8% (at 0.5 mg/L)	ISO 9297
Sulfate	mg/L	3	10% (in a range of 5-60 mg/L)	VL-ANP-17
Fluoride	mg/L	0.02	8% (at 0.5 mg/L)	ISO 10359-1

Trace elements in filtrated water samples (0.45  $\mu\text{m}$ ) were analyzed in the laboratory of GBA Gesellschaft für Bioanalytik mbH (Pinneberg, Germany) by following accredited methods (see Table 4, Appendix 1).

Table 4 Selected trace elements analyzed in surface water samples in the laboratory of GBA.

Element	Unit	Limit of quantification	Uncertainty (%)	Method
Mercury, Hg	mg/L	0.00020	4.8	EN ISO 17294



Element	Unit	Limit of quantification	Uncertainty (%)	Method
Cadmium, Cd	mg/L	0.00030	5.3	EN ISO 17294
Lead, Pb	mg/L	0.0010	4.8	EN ISO 17294
Nickel, Ni	mg/L	0.0010	6.5	EN ISO 17294
Antimony, Sb	mg/L	0.0010	nd	EN ISO 17294
Arsenic, As	mg/L	0.00050	5.3	EN ISO 17294
Barium, Ba	mg/L	0.00050	nd	EN ISO 17294
Chromium, Cr total	mg/L	0.0010	7.2	EN ISO 17294
Copper, Cu	mg/L	0.0010	6.9	EN ISO 17294
Molybdenum, Mo	mg/L	0.0010	nd	EN ISO 17294
Vanadium, V	mg/L	0.0010	nd	EN ISO 17294
Zinc, Zn	mg/L	0.0050	8.4	EN ISO 17294

nd – no data.

### 3.5. Legal limits

Results of the analysis were compared against environmental quality standards. Among measured parameters there are no limit value for the temperature, electric conductivity, content of chloride and sulfate as well as content of following trace elements such as vanadium (V), antimony (Sb) and molybdenum (Mo) in surface water according to Estonian legal regulations. For these parameters the limit values for drinking and groundwater were used as indicative values (except V and Sb).

Following regulations were used:

#### Selected trace elements in soil samples:(Table 5):

Regulation No 38 of the Estonian Ministry of Environment (adopted in 11.08.2010), Ohtlike ainete sisalduse piirväärtused pinnases (*Concentration limits of hazardous substances in the soil*), Riigi Teataja, I 2010, 57, 373 (in Estonian).

Table 5 Target and permitted values of selected trace elements in soil.

Element	Unit	Target value*	Permitted value** in residential area	Permitted value** in industrial area
As	mg/kg	20	30	50
Pb	mg/kg	50	300	600
Cd	mg/kg	1	5	20
Cr sum	mg/kg	100	300	800
Cu	mg/kg	100	150	500
Ni	mg/kg	50	150	500
Hg	mg/kg	0.5	2	10
Zn	mg/kg	200	500	1000
Sb	mg/kg	10	20	100
Ba	mg/kg	500	750	2000
Mo	mg/kg	10	20	200
V	mg/kg	50	300	1000

\* target value shows the content below which the soil status is considered good.

\*\* permitted value shows the content above which the soil is considered contaminated.

**Electric conductivity, chloride and sulfate content** in surface water are not regulated. Therefore the regulation on drinking water quality was used for indicative purposes (Table 6):

Regulation No 82 of the Ministry of Social Affairs (adopted 31.07.2001), *Joogivee kvaliteedi- ja kontrollinõuded ning analüüsimeetodid (Quality and control requirements for drinking water and methods of analysis)*, Riigi Teataja I, 11.01.2013, 2 (in Estonian).

Table 6 Indication of limit values in drinking water.

Parameter	Unit	Limit value
Electric conductivity	µS/cm	2500
Temperature	°C	-
Chloride	mg/L/	250
Sulfate	mg/L	250

**pH** in surface water has to be in a range of 6-9 in order to consider the status of the water body to be good:

Regulation No 44 of the Estonian Ministry of Environment (adopted in 28.07.2009), *Pinnaveekogumite moodustamise kord ja nende pinnaveekogumite nimestik, mille seisundiklass tuleb määrata, pinnaveekogumite seisundiklassid ja seisundiklassidele vastavad kvaliteedinäitajate väärtused ning seisundiklasside määramise kord (The procedure for the formation of surface water bodies and the list of these surface water bodies, which class status shall be determined, the status of surface water bodies and the quality indicators for the status of the corresponding water bodies and the procedure for determining the status)*, Riigi Teataja I, 25.11.2010, 15 (in Estonian)

**Content of priority substances in surface water samples** (Table 7):

Directive 2013/39/EU of the European Parliament and of the Council of 12 August 2013 amending Directives 2000/60/EC and 2008/105/EC as regards priority substances in the field of water policy..

Table 7 Environmental quality standards of priority hazardous metals in surface water.

Name of substance	Unit	AA-EQS*	MAC-EQS**
Hg	mg/L		0.00007
Cd**	mg/L	0.00015	0.0009
Pb	mg/L	0.0012****	0.014
Ni	mg/L	0.004****	0.034

\* annual average value of the environmental quality standard;

\*\* maximum allowable concentration of the environmental quality standard;

\*\*\* For Cd the EQS values vary depending on the hardness of the water. Hardness of the water was considered 100 to < 200 mg CaCO<sub>3</sub>/L – class 4 [7];

\*\*\*\* bioavailable concentration.

**Content of selected hazardous substances in surface water samples (Table 8):**

Regulation No 49 of the Estonian Ministry of Environment (adopted in 09.09.2010), Pinnavee keskkonna kvaliteedi piirväärtused ja nende kohaldamise meetodid ning keskkonna kvaliteedi piirväärtused vee-elustikus (*The environmental quality limit values for the surface water and their methods of application, and the environmental quality limit values for water biota*), Riigi Teataja I, 18.12.2013, 5 (in Estonian).

Table 8 Environmental quality standards of hazardous substances in surface water.

Name of substance	Unit	MAC-EQS
Fluoride	mg/L	1.5
As	mg/L	0.01
Ba	mg/L	0.1*
Cr total	mg/L	0.005
Cu	mg/L	0.015

\* in amendment, expected to be in force since January 2015

**Content of molybdenum** in surface water samples was indicatively compared to limit value in groundwater :0.07 mg/L.

Regulation No 39 of the Estonian Ministry of Environment (adopted in 11.08.2010), Ohtlike ainete põhjavee kvaliteedi piirväärtused (*The environmental quality limit values of the hazardous substances in groundwater*)

**3.6.Previous monitoring**

Whenever possible the results of previous monitoring programs were used to obtain natural background concentrations and to analyze the trends in contents of selected parameters in soil and surface water samples.

Previous soil sampling episodes have taken place in Narva-Mustajõe pilot section in years 2011 and 2013 and in Simuna-Vaiatu section in 2012 (Figure 4).



Figure 4. Soil sampling points in previous and current monitoring programs in Narva-Mustajõe (left) and Simuna-Vaiatu (right) pilot sections.

Table 9 presents the results of average contents of selected elements in composite samples of Narva-Mustajõe and Simuna-Vaiatu pilot sections published in monitoring program carried out in 2012 [8]. The results can be considered as a natural background content of selected elements in the soil of pilot sections.

Table 9 Content of selected trace elements in soil samples of the pilot sections presented in previous monitoring report [8].

Date		30.05.2012	30.05.2012
Pilot section		Narva-Mustajõe	Simuna-Vaiatu
Element	Unit		
As	mg/kg	4.6	3.5
Pb	mg/kg	13	15
Cd	mg/kg	<0.2	0.26
Cr sum	mg/kg	18	7.3
Cu	mg/kg	<10	<10
Ni	mg/kg	10	4.1
Hg	mg/kg	<0.1	<0.1
Zn	mg/kg	40	24
Sb	mg/kg	<0.5	<0.5
Ba	mg/kg	76	65
Mo	mg/kg	<2	<2
V	mg/kg	34	11
Se	mg/kg	<1	<1

Surface water sampling has been previously carried out in Narva-Mustajõe and Simuna-Vaiatu pilot sections since 2011 (see Figure 2). It must be noted that the list of trace elements analyzed in previous monitoring programs is shorter than in follow-up program and the samples were not filtrated before trace element analyzes. The results of previous sampling episodes are presented in Table 10 [8]. The results from year 2011 can be considered as a natural background of surface water in an area.

Table 10. Results of previous surface water monitoring program [8].

Parameter	Unit	Narva-Mustajõe				Simuna-Vaiatu			
		27.07.11	26.07.12	17.10.12	30.07.13	27.07.2011		18.11.2013	
pH		7.41	8.3	7.6	7.9	7.86	7.29	6.9	7.1
EC	µS/cm	1034	702	763	725	299	571	74	257
Chloride	mg/L	44	78	24	39	3.5	7	<3	<3
Sulfate	mg/L	<5	22	74	31	<5	<5	<5	<5
As	mg/L	0.0081	0.00073	0.00047	0.00062	0.0006	0.0011	0.00066	0.00054
Cr	mg/L	0.00078	<0.0005	<0.0005	0.00076	<0.0005	<0.0005	<0.0005	<0.0005
Pb	mg/L	0.0077	0.0018	<0.0001	0.00063	0.00018	0.011	0.00057	<0.0005
Mo	mg/L	0.00066	0.00057	0.00077	0.0012	0.00013	0.00015	<0.0001	<0.0001
V	mg/L	0.0013	<0.0005	<0.0005	<0.0005	<0.0005	0.0017	<0.0005	<0.0005

## **4. Results**

The results of analysis of soil and surface water samples taken in 2014 and 2015 are presented in following Table 11-Table 18.

### 3.7. Soil

Table 11. Contents of selected elements in Narva-Mustajõe soil samples (locations see Figure 1).

Date			30.05.2012	22.07.2014			22.07.2015		
Sample name			Composite	NM2014-1	NM2014-2	NM2014-3	NM2015-1	NM2015-2	NM2015-3
Parameter	Unit								
Sampling depth	m		0-0.3	0.2-0.4	0.2-0.4	0.2-0.4	0.05-0.2	0.05-0.2	0.05-0.2
Dry weight	mass-%	-	83	82.8	92.2	95	92.1	86	92.1
Element		TV*	NBC**						
Arsenic, As	mg/kg	20	4.6	11	4.7	2.4	3.2	4	1.6
Lead, Pb	mg/kg	50	13	14	13	6.9	13	11	5.9
Cadmium, Cd	mg/kg	1	<0.2	0.11	<0.1	<0.1	0.12	<0.1	<0.1
Chrome, Cr sum	mg/kg	100	18	24	15	7.6	9	8	5.2
Copper, Cu	mg/kg	100	<10	15	11	6.3	8.6	9.9	4.8
Nickel, Ni	mg/kg	50	10	14	9.8	4	5.6	6.8	2.8
Mercury, Hg	mg/kg	0.5	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Zinc, Zn	mg/kg	200	40	55	39	25	36	46	25
Antimony, Sb	mg/kg	10	<0.5	<0.1	<0.1	<0.1	<1.0	<1	<1
Barium, Ba	mg/kg	500	76	168	59	31	37	51	19
Molybdenum, Mo	mg/kg	10	<2	2.3	<0.1	<0.1	<1	<1	<1
Vanadium, V	mg/kg	50	34	31	26	10	15	14	7.5
Selenium, Se	mg/kg	1	<1	<2.0	<2.0	<2.0	<2.0	<2.0	<2.0

\* Target value, see p. 3.5

\*\* Natural background content, see p. 3.6

Table 12 Contents of selected elements in Simuna-Vaiatu soil samples (locations see Figure 1).

Date			30.05.2012	22.07.2014		22.07.2015	
Sample name			Composite	SV2014-2	SV2014-3	SV2015-2	SV2015-3
Parameter	Unit						
Depth	m		0-0.3	0.2-0.4	0.2-0.4	0.05-0.2	0.05-0.2
Dry weight	mass-%		26	82.3	88.2	53.2	86.3
Element		TV*	NBC**				
As	mg/kg	20	3.5	2.3	2	6.3	2.7
Pb	mg/kg	50	15	4.6	5.7	13	7.1
Cd	mg/kg	1	0.26	<0.1	<0.1	0.1	<0.1
Cr sum	mg/kg	100	7.3	6.8	5.7	9.3	4.9
Cu	mg/kg	100	<10	4.4	4.4	6	5.2
Ni	mg/kg	50	4.1	3.7	3.3	4.9	3.1
Hg	mg/kg	0.5	<0.1	<0.1	<0.1	<0.1	<0.1
Zn	mg/kg	200	24	24	22	32	24
Sb	mg/kg	10	<0.5	<0.1	<0.1	<1	<1
Ba	mg/kg	500	65	24	20	60	20
Mo	mg/kg	10	<2	<0.1	<0.1	1.2	<1
V	mg/kg	50	11	8.9	9.8	11	7.7
Se	mg/kg	1	<1	<2.0	<2.0	<2.0	<2.0

\* Target value, see p. 3.5

\*\* Natural background content, see p. 3.6



### 3.8.Surface water

Table 13 Indicative parameters of surface water samples from Narva-Mustajõe pilot section in 2014 and 2015, comparison with indicators and natural background level. Sampling site locations are presented in Figure 2.

Sample name				NM2014-1			NM2014-2			NM2014-3		
Date		27.07.11		27.05	30.07	22.10	27.05	30.07	22.10	27.05	30.07	22.10
Parameter	Unit	Indicative value	Natural background									
pH		range 6-9	7.41	7.48	7.93	7.26	6.94	7.5	7.48	7.96	7.56	7.66
Electric conductivity	µS/cm	max 2500	1034	522	603	969	931	650	683	1012	328	481
Chloride	mg/L/	max 250	44	11	17	28	19	65	43	63	22	29
Sulfate	mg/L	max 250	<5	23	<3	126	22	7	48	23	39	29
Fluoride	mg/L	max 1.5	nd	0.27	0.34	0.30	0.26	0.17	0.13	0.16	0.22	0.13

Sample name				NM2015-1					NM2015-2					NM2015-3				
Date		27.07.2011		3.02	14.04	21.07	20.10	24.11	3.02	14.04	21.07	20.10	24.11	3.02	14.04	21.07	20.10	24.11
Parameter.	Unit	Indic. value	Nat. back-ground															
pH		range 6-9	7.41	7.16	7.98	7.56	7.41	7.01	7.69	7.9	7.41	7.64	7.53	7.68	7.88	8.01	7.63	7.44
EC	µS/cm	max 2500	1034	996	831	537	1277	712	887	973	1523	1488	536	908	939	398	1055	518
Chloride	mg/L/	max 250	44	13	17	5.4	20	25	60	68	<b>295</b>	135	27	51	65	12	91	64
Sulfate	mg/L	max 250	<5	43	81	<3	<b>525</b>	<b>297</b>	39	39	35	182	211	32	39	6	76	52
Fluoride	mg/L	max 1.5	nd	0.25	0.3	0.3	0.34	0.26	0.13	0.13	0.15	0.14	0.15	0.13	0.13	0.14	0.16	0.2

nd – no data

Table 14 Indicative parameters of surface water samples from Simuna-Vaiatu pilot section in 2014 and 2015, comparison with indicators and natural background level. Sampling site locations are presented in Figure 2.

Sample name				SV2014-1			SV2014-2			SV2014-3		
Date			27.07.11	27.05.	30.07.	22.10.	27.05.	30.07.	22.10.	27.05.	30.07.	22.10.
Parameter	Unit	Indicative value	Natural background									
pH		range 6-9	7.58	8.06	7.79	7.72	6.94	7.14	7.54	8.02	8.21	7.95
Electric conductivity	µS/cm	max 2500	435	509	560	881	931	178	252	215	663	273
Chloride	mg/L/	max 250	5.3	9	5	8	19	<3	<3	<3	10	<3
Sulfate	mg/L	max 250	<5	22	65	162	22	<3	<3	<3	118	10
Fluoride	mg/L	max 1.5	nd	0.22	0.24	0.20	0.06	0.17	0.08	0.07	0.43	0.09

Sample name				SV2015-1				SV2015-2				SV2015-3			
Date			27.07.11	03.02.	14.04	21.07.	20.10.	03.02.	14.04	21.07.	20.10.	03.02.	14.04	21.07.	20.10.
Parameter	Unit	Indicative value	Natural background												
pH		range 6-9	7.58	7.27	8.01	8.02	8.28	7.29	7.46	8	7.78	7.32	7.72	8.37	7.47
Electric conductivity	µS/cm	max 2500	435	585	522	637	496	226	218	633	598	245	252	469	646
Chloride	mg/L/	max 250	5.3	3	<3	<3	3.2	3	<3	12	12	<3	<3	7.7	11
Sulfate	mg/L	max 250	<5	42	36	55	21	<3	<3	43	25	<3	<3	29	27
Fluoride	mg/L	max 1.5	nd	0.10	0.12	0.26	0.20	0.07	0.07	0.17	0.12	0.08	0.08	0.18	0.13

Table 15. Trace elements in filtrated (0.45 µm) surface water samples from Narva-Mustajõe pilot section in 2014 and comparison with EQS and available background level.

Sample name				NM2014-1			NM2014-2			NM2014-3		
Date			27.07.11	27.05.	30.07	22.10.	27.05.	30.07.	22.10.	27.05.	30.07.	22.10.
Element	Unit	MAC-EQS*	Natural background**									
Hg	mg/L	0.00007	nd	<0.00020	<0.00020	<0.00020	<0.00020	<0.00020	<0.00020	<0.00020	<0.00020	<0.00020
Cd	mg/L	0.0009	nd	<0.00030	<0.00030	<0.00030	<0.00030	<0.00030	<0.00030	<0.00030	<0.00030	<0.00030
Pb	mg/L	0.014	0.0077	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010
Ni	mg/L	0.034	nd	0.0014	<0.0010	<0.0010	0.0041	<0.0010	<0.0010	0.0021	0.0013	<0.0010
As	mg/L	0.01	0.0081	0.00064	0.0013	<0.00050	0.0012	0.0012	0.00058	0.0016	0.0011	<0.00050
Ba	mg/L	0.1	<b>0.36***</b>	<b>0.21</b>	<b>0.29</b>	<b>0.36</b>	<b>0.19</b>	0.078	<b>0.15</b>	0.049	0.024	0.09
Cr sum	mg/L	0.005	0.78	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
Cu	mg/L	0.015	nd	<0.001	<0.001	<0.001	0.0048	<0.001	<0.001	0.0011	0.0026	0.0018
Zn	mg/L	0.01	nd	<0.0050	0.0061	<0.0050	0.012	0.0076	<0.0050	<0.0050	<0.0050	<0.0050
Mo	mg/L		0.00066	<0.0010	<0.0010	<0.0010	0.0016	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010
Sb	mg/L		nd	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010
V	mg/L		0.0013	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010

\* MAC-EQS for priority substances Directive 2013/39/EU, other hazardous substances Regulation No 49 of the Estonian Ministry of Environment;

\*\*2011 sampling episode (non-filtrated sample);

\*\*\* Sample NM-0 taken in 21.07.2015 for natural background level outside the pilot section area

Table 16. Trace elements in filtrated (0.45 µm) surface water samples from Narva-Mustajõe pilot section in 2015 and comparison with EQS and available background level.

Sample name				NM2015-1				NM2015-2				NM2015-3			
Date		27.07.11		03.02.	14.04	21.07.	20.10.	03.02.	14.04	21.07.	20.10.	03.02.	14.04	21.07.	20.10.
Element	Unit	MAC-EQS*	Natural background**												
Hg	mg/L	0.00007	nd	<0.0002	<0.00020	<0.00020	<0.0002	<0.0002	<0.00020	<0.00020	<0.0002	<0.0002	<0.00020	<0.00020	<0.0002
Cd	mg/L	0.0009	nd	<0.0003	<0.00030	<0.00030	<0.0003	<0.0003	<0.00030	<0.00030	<0.0003	<0.0003	<0.00030	<0.00030	<0.0003
Pb	mg/L	0.014	0.0077	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010
Ni	mg/L	0.034	nd	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010
As	mg/L	0.01	0.0081	0.00057	<0.00050	0.0008	0.0005	<0.0005	<0.00050	<0.00050	0.00051	<0.0005	<0.00050	0.00063	0.00055
Ba	mg/L	0.1	<b>0.36***</b>	<b>0.4</b>	<b>0.3</b>	<b>0.3</b>	<b>0.18</b>	<b>0.17</b>	<b>0.19</b>	<b>0.32</b>	<b>0.14</b>	<b>0.2</b>	<b>0.18</b>	<b>0.14</b>	<b>0.19</b>
Cr sum	mg/L	0.005	0.78	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.001	<0.001	<0.001	<0.0010
Cu	mg/L	0.015	nd	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	0.0022	<0.001	<0.001	0.0011	<0.0010
Zn	mg/L	0.01	nd	<0.0050	<0.0050	<0.0050	<0.0050	<0.0050	<0.0050	<0.0050	<0.0050	<0.0050	<0.0050	<0.0050	<0.0050
Mo	mg/L		0.00066	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010
Sb	mg/L		nd	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010
V	mg/L		0.0013	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010

\* MAC-EQS for priority substances Directive 2013/39/EU, other hazardous substances Regulation No 49 of the Estonian Ministry of Environment;

\*\*2011 sampling episode (non-filtrated sample);

\*\*\* Sample NM-0 taken in 21.07.2015 for natural background level outside the pilot section area

Table 17 Trace elements in filtrated (0.45 µm) surface water samples from Simuna-Vaiatu pilot section in 2014 and comparison with EQS and available background level.

Sample name				SV2014-1			SV2014-2			SV2014-3		
Date		27.07.11		27.05.	30.07	22.10.	27.05.	30.07.	22.10.	27.05.	30.07.	22.10.
Element	Unit	MAC-EQS*	Natural background**									
Hg	mg/L	0.00007	nd	<0.00020	<0.00020	<0.00020	<0.00020	<0.00020	<0.00020	<0.00020	<0.00020	<0.00020
Cd	mg/L	0.0009	nd	<0.00030	<0.00030	<0.00030	<0.00030	<0.00030	<0.00030	<0.00030	<0.00030	<0.00030
Pb	mg/L	0.014	0.0056	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010
Ni	mg/L	0.034	nd	0.0032	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	0.0014	<0.0010
As	mg/L	0.01	0.0009	0.0045	0.0037	0.0014	0.00071	0.0027	0.00068	0.0010	0.0041	0.00072
Ba	mg/L	0.1	nd	0.022	0.032	0.053	0.015	0.049	0.017	0.017	0.045	0.019
Cr sum	mg/L	0.005	<0.0005	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
Cu	mg/L	0.015	nd	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
Zn	mg/L	0.01	nd	<0.0050	0.0061	<0.0050	<0.0050	0.0076	0.0058	<0.0050	<0.0050	0.0056
Mo	mg/L		0.00014	0.0041	0.0057	0.0044	<0.0010	0.0022	<0.0010	<0.0010	0.0074	<0.0010
Sb	mg/L		nd	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010
V	mg/L		0.0011	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010

\* MAC-EQS for priority substances Directive 2013/39/EU, other hazardous substances Regulation No 49 of the Estonian Ministry of Environment;

\*\*2011 sampling episode (non-filtrated sample);

Table 18. Trace elements in filtrated (0.45 µm) surface water samples from Simuna-Vaiatu pilot section in 2015 and comparison with EQS and available background level

Sample name				SV2014-1				SV2014-2				SV2014-3			
Date		27.07.11		03.02.	14.04	21.07.	20.10.	03.02.	14.04	21.07.	20.10.	03.02.	14.04	21.07.	20.10.
Element	Unit	MAC-EQS*	Natural back-ground**												
Hg	mg/L	0.00007	nd	<0.0002	<0.0002	<0.0002	<0.0002	<0.0002	<0.0002	<0.0002	<0.0002	<0.0002	<0.0002	<0.0002	<0.0002
Cd	mg/L	0.0009	nd	<0.0003	<0.0003	<0.0003	<0.0003	<0.0003	<0.0003	<0.0003	<0.0003	<0.0003	<0.0003	<0.0003	<0.0003
Pb	mg/L	0.014	0.0056	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010
Ni	mg/L	0.034	nd	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010
As	mg/L	0.01	0.0009	0.00066	0.00052	0.0032	0.001	0.00072	<0.0005	0.00062	<0.0005	0.00074	0.00068	0.001	<0.0005
Ba	mg/L	0.05	nd	0.036	0.031	0.05	0.0097	0.015	0.014	0.14	0.095	0.016	0.018	0.037	<b>0.14</b>
Cr sum	mg/L	0.005	<0.0005	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010
Cu	mg/L	0.015	nd	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010
Zn	mg/L	0.01	nd	<0.0050	<0.0050	<0.0050	0.0065	<0.0050	<0.0050	<0.0050	<0.0050	<0.0050	<0.0050	<0.0050	<0.0050
Mo	mg/L		0.00014	<0.0010	0.0013	0.0077	0.001	<0.0010	<0.0010	0.0029	<0.0010	<0.0010	<0.0010	0.0011	<0.0010
Sb	mg/L		nd	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010
V	mg/L		0.0011	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010

\* MAC-EQS for priority substances Directive 2013/39/EU, other hazardous substances Regulation No 49 of the Estonian Ministry of Environment;

\*\*2011 sampling episode (non-filtrated sample);

## 5. Summary of the results and trends

During the course of the environmental monitoring in 2014 and 2015 following sampling campaigns were conducted in both of the pilot sections of Narva-Mustajõe and Simuna-Vaiatu roads:

Two soil sampling episodes and seven surface sampling episodes (see p. 3.1). In addition two surface water sampling episodes in Narva Mustajõe pilot section were conducted in 2015 for checking the sulfate content and natural background level of barium. Sampling reports as well as results of analysis are presented in Appendix 2.

### 5.2 Soil

Soil samples were taken from the banks of both pilot road sections at depth 0.2-0.4 m in 2014 and 0.05-0.2 in 2015. The content of trace elements in soil samples was compared to the legal limits and natural background level. Content of all trace elements was below target values set by the national regulation<sup>1</sup> in all samples.

In Simuna-Vaiatu road previous soil sampling was conducted in 30.05.2012. In following table the content of trace elements in composite sample taken in 2012 and mean content of elements in the soil samples taken in 2014 and 2015 is compared (Table 19 and Table 20). For the sake of clarity also the mean content of the trace elements in Estonian soil is presented.

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<sup>1</sup> No 38 of the Estonian Ministry of Environment (adopted in 11.08.2010), Ohtlike ainete sisalduse piirväärtused pinnases (Concentration limits of hazardous substances in the soil), Riigi Teataja, I 2010, 57, 373 (In Estonian)

Table 19. Average content of selected elements (mg/kg<sub>dry</sub>) in Estonian soil according to literature compared to average content in Narva-Mustajõe soil samples.

Element	Estonian soil mean <sup>2</sup>	Narva-Mustajõe composite 30.05.12	Current monitoring	
			Narva-Mustajõe mean (n=3) 22.07.14	Narva-Mustajõe mean (n=3) 22.07.15
As	nd.	4.6	6.0	2.9
Pb	16	13	11.3	10.0
Cd	0.4	<0.2	0.1	0.12
Cr <sub>sum</sub>	42	18	15.5	7.4
Cu	11	<10	10.8	7.8
Ni	22	10	9.3	5.1
Hg	0.03	<0.1	<0.1	<0.1
Zn	37	40	39.7	35.7
Sb	nd.	<0.5	<0.1	<0.1
Ba	383	76	86	35.8
Mo	1.3	<2	2.3	<1.0
V	44	34	22.3	12.2
Se	nd	<1	<2.0	<2.0

nd – no data.

In Narva-Mustajõe pilot section the content of selected elements in composite sample taken in 2012 and mean value in samples taken in 2014 and 2015 is in the same order of magnitude. Only the mean content of Ba and Mo is over 10% higher in samples taken in 2014 compared to 2012. At the same time the content of Ba in soil samples taken in 2015 is almost two times lower compared to 2012.

Table 20. Average content and range of selected elements (mg/kg<sub>dry</sub>) in Estonian soil compared to average content in Simuna-Vaiatu soil samples.

Element	Estonian soil mean	Simuna-Vaiatu composite 30.05.12	Simuna-Vaiatu mean (n=2) 22.07.14	Simuna-Vaiatu mean (n=2) 22.07.15
As	nd.	3.5	2.2	4.5
Pb	16	15	5.2	10.1
Cd	0.4	0.26	<0.1	0.08
Cr <sub>sum</sub>	42	7.3	6.3	7.1
Cu	11	<10	4.4	5.6
Ni	22	4.1	3.5	4
Hg	0.03	<0.1	<0.1	<0.1
Zn	37	24	23	28
Sb	nd	<0.5	<0.1	<1.0
Ba	383	65	22	40
Mo	1.3	<2	<0.1	0.6
V	44	11	9.4	9.4
Se	nd	<1	<2.0	<2.0

<sup>2</sup> V. Petersell, H. Ressar, M. Carlson, V. Mõttus, M. Enel, A. Mardla, K. Täht, The geochemical atlas of the humus horizon of Estonian soil, Eesti Geoloogiakeskus, 1997.



As can be seen from the table the mean content of almost all analyzed trace elements lower in samples taken in 2014 and 2015 compared to composite sample taken in 2012. Exceptions are arsenic and zinc – their content in 2015 samples is less than 10 per cent higher compared to 2012 samples. It must still be noted that the dry weight of one sample taken in 2015 at Simuna-Vaiatu pilot section was 53.2 wt-%, which makes the sample relatively wet.

When compared the content of soil in current follow-up monitoring program to previous monitoring and to average content in Estonian soil following conclusions can be drawn:

- the effect of OSA in road construction onto content of trace elements in soil is negligible.

## **5.2 Surface water**

For following the long-term trends (2011-2015) of selected parameters in surface water in the pilot areas it is possible to use only those parameters that were measured in previous monitoring campaigns i.e. pH, EC, chloride, sulfate, As, Cr, Pb, Mo, V. Content of Cr and V in all water samples is below limits of detection therefore these metals are not included in the analysis. For the calculation of average concentrations, values below the limit of quantification were set to half of the value of the limit of quantification concerned [9].

### **5.2.1 Indicative parameters**

Following indicative parameters are presented pH, electric conductivity, content of chlorides and sulfates (Figure 5-Figure 8). These indicative parameters were also monitored in previous environmental programs of two pilot sections.

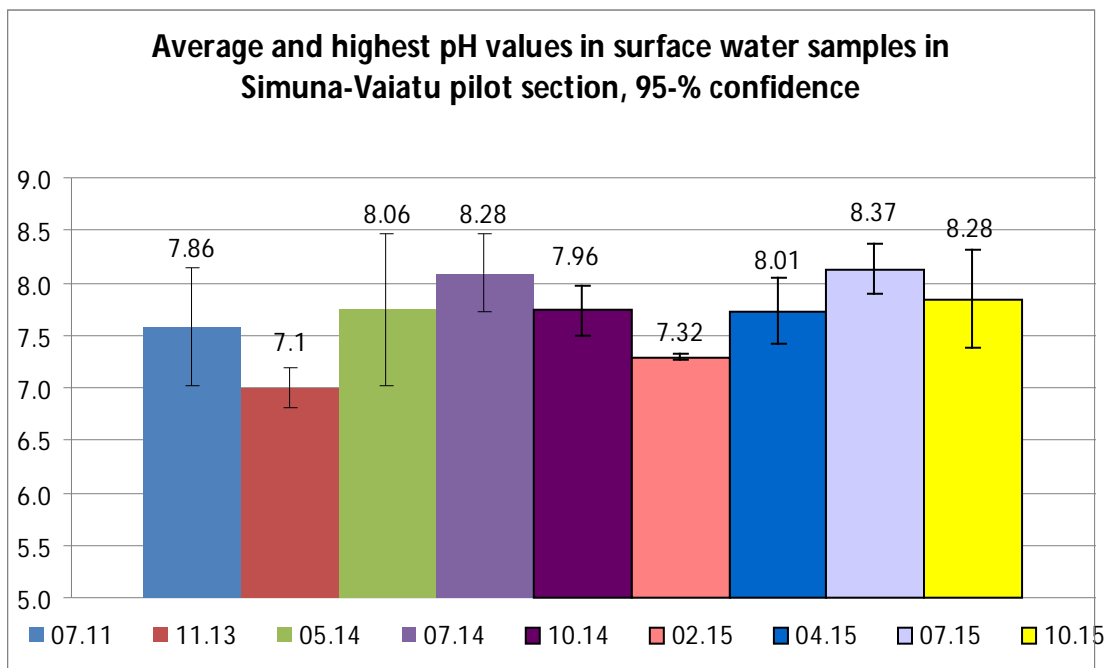
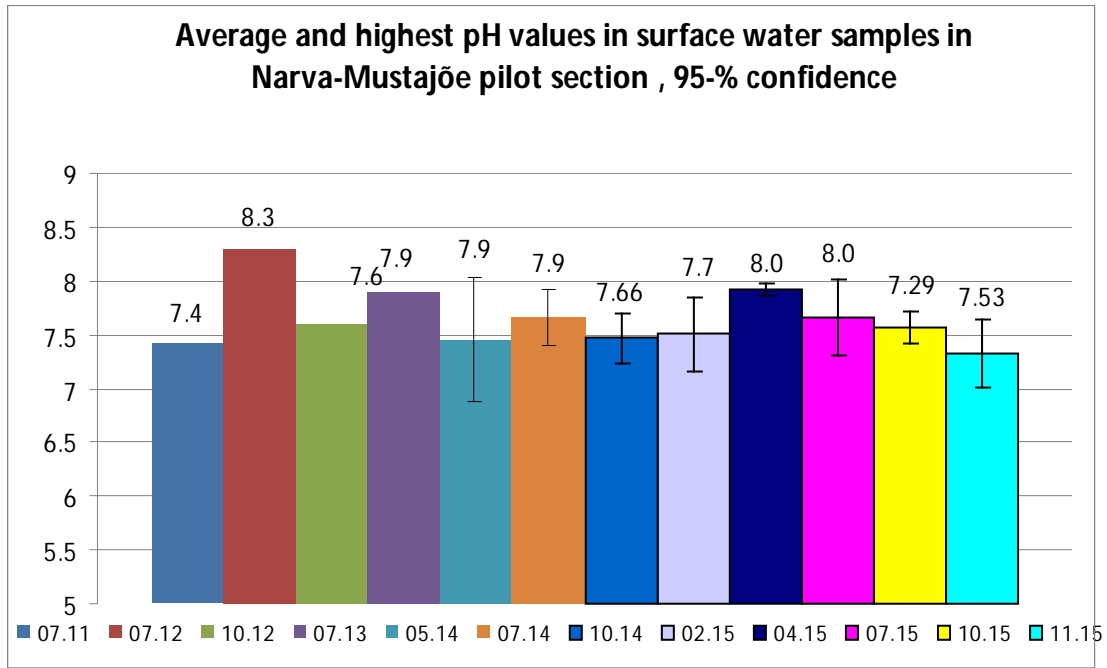


Figure 5 Observed pH values of water samples during monitoring of Narva-Mustajõe and Simuna-Vaiatu pilot sections.

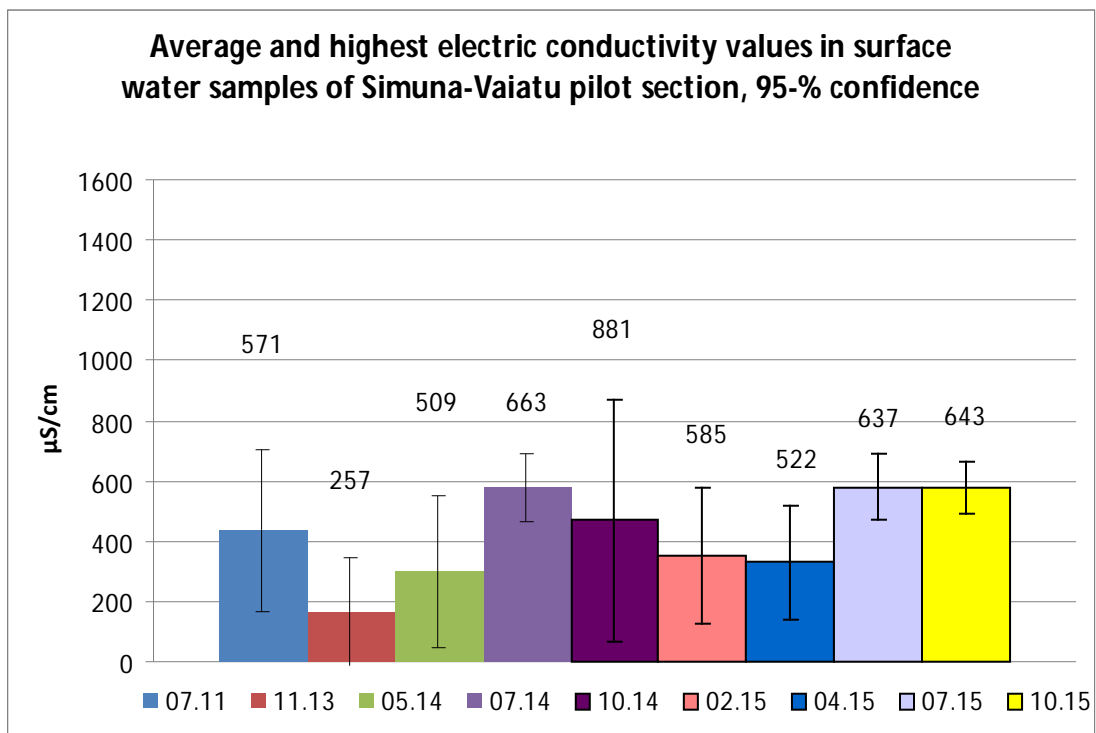
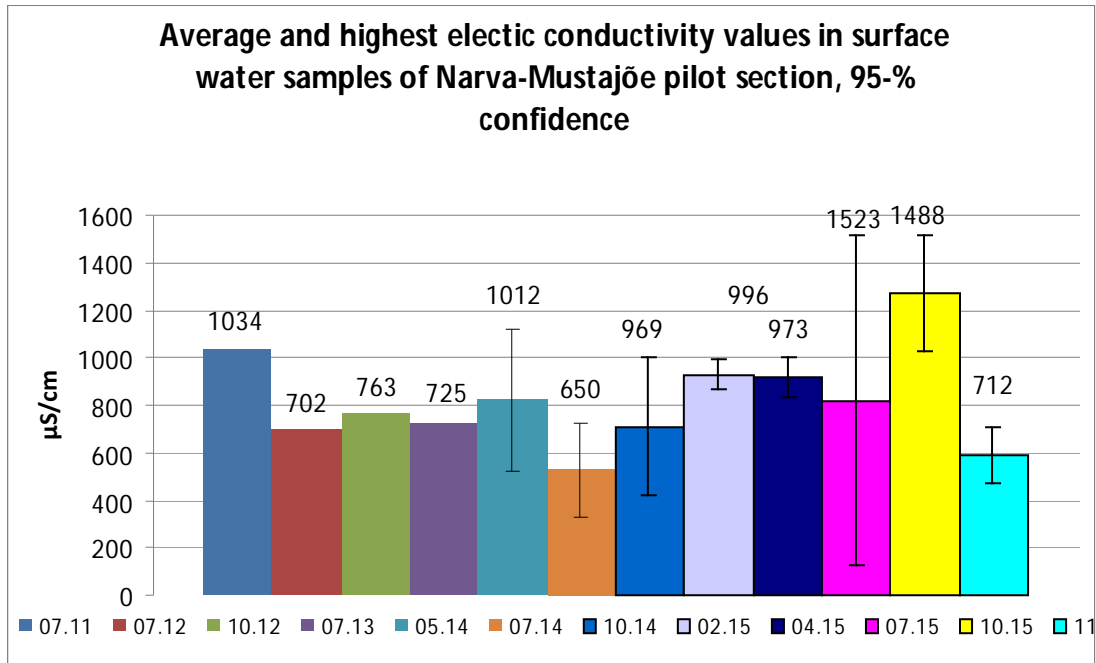


Figure 6 Observed electric conductivity values of water samples of Narva-Mustajõe and Simuna-Vaiatu pilot sections.

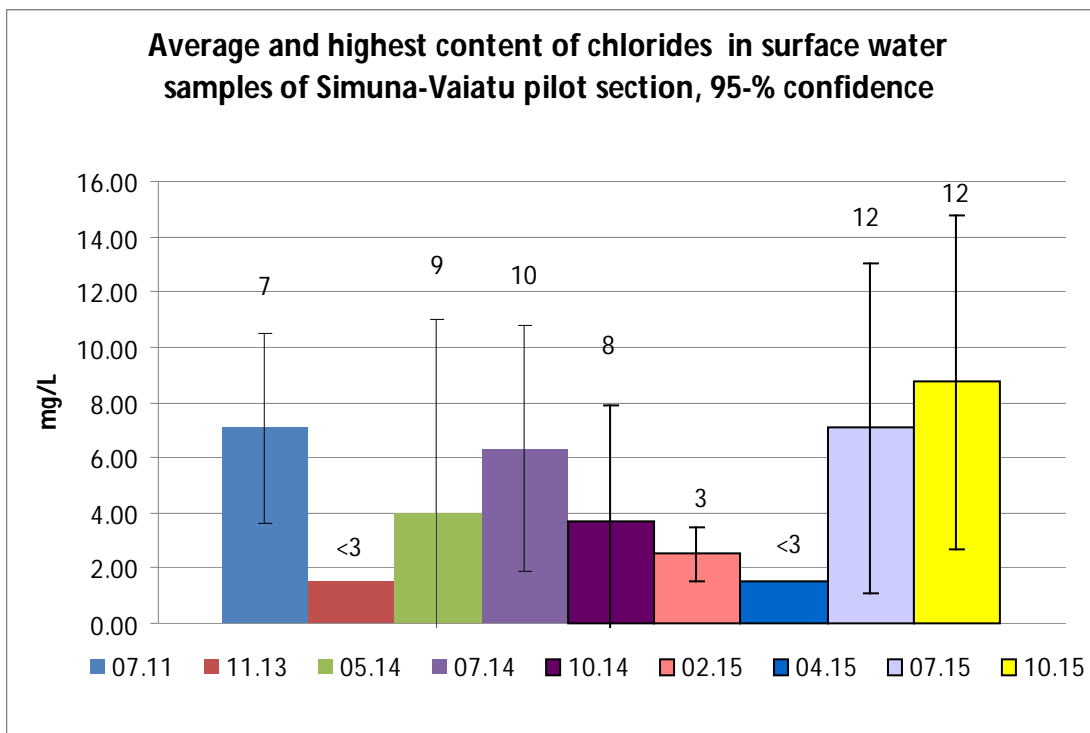
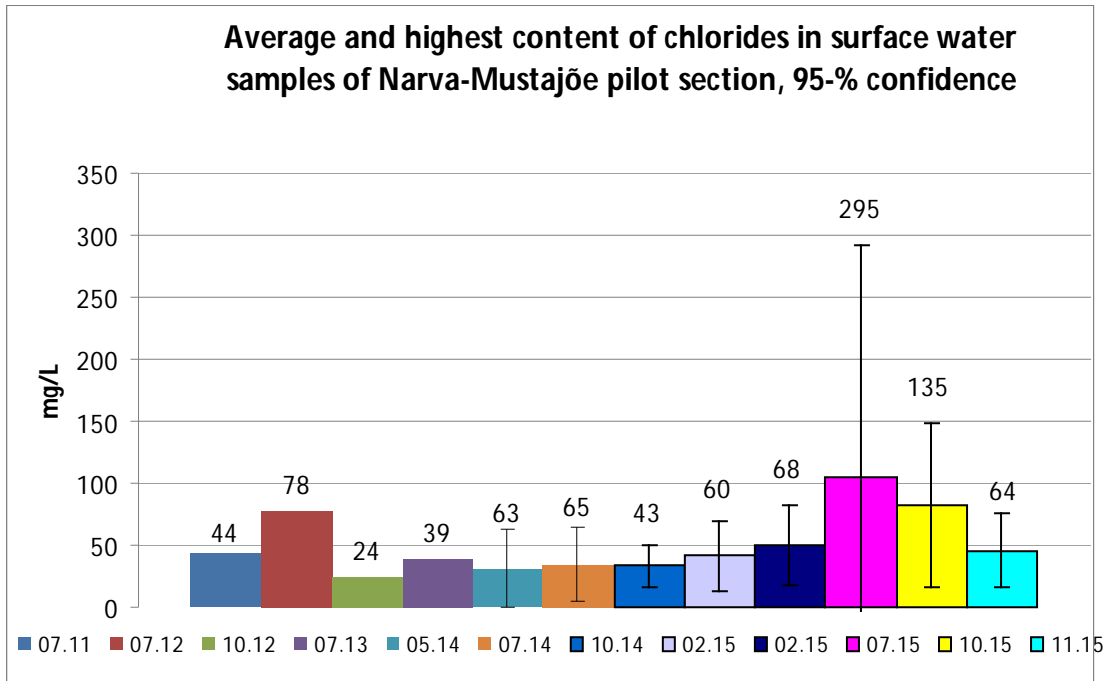


Figure 7 Observed content of chlorides in water samples of Narva-Mustajõe and Simuna-Vaiatu pilot sections. NB! Different scales.

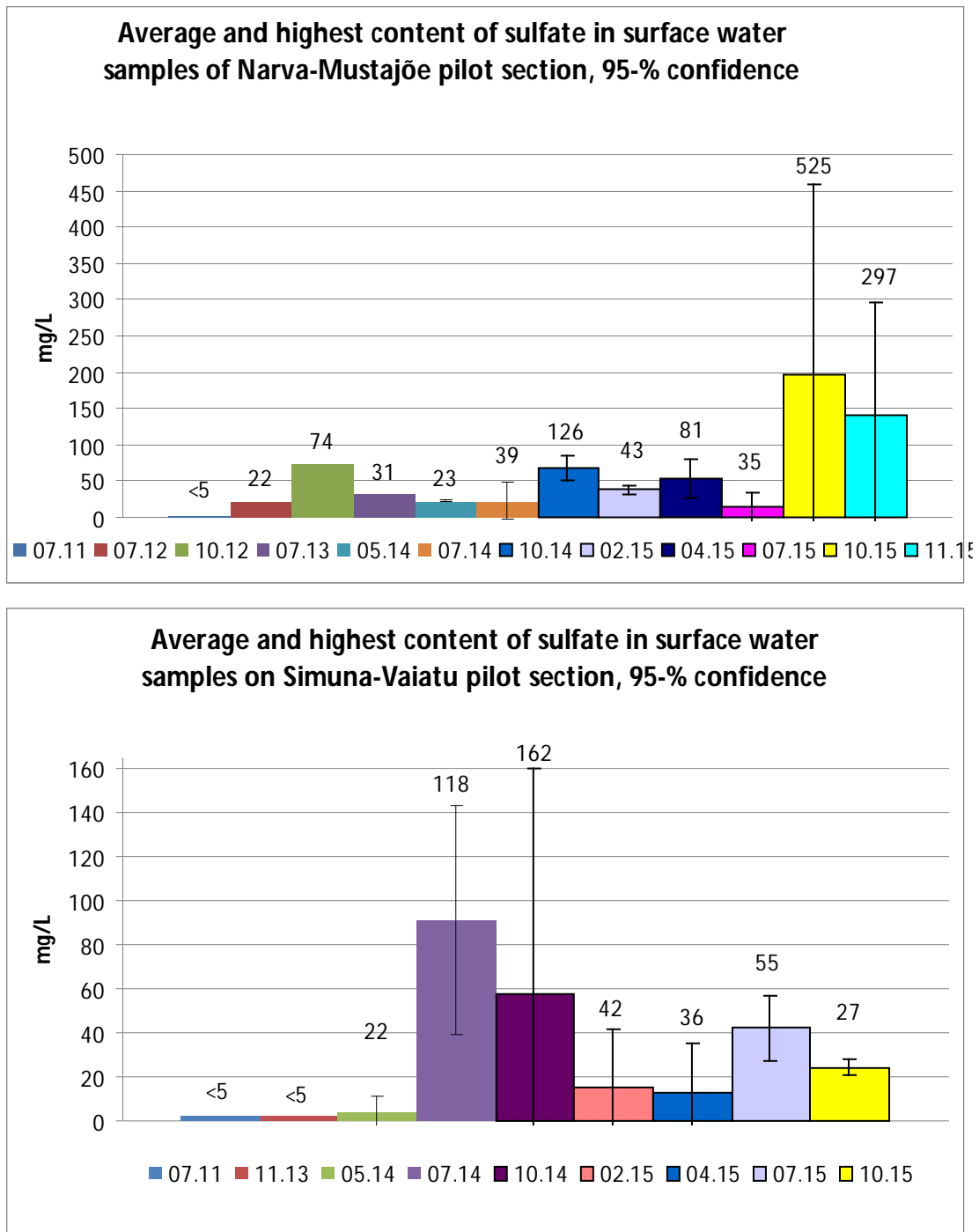


Figure 8 Observed content of sulfate in water samples of Narva-Mustajõe and Simuna-Vaiatu pilot sections. NB! Different scales.

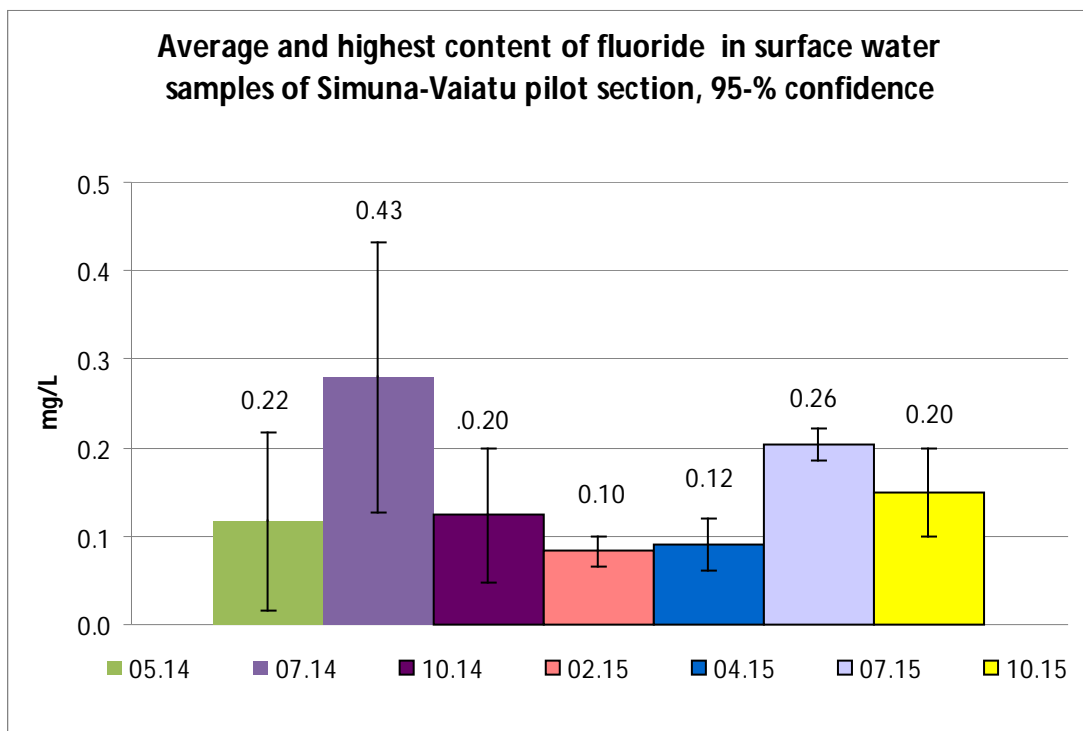
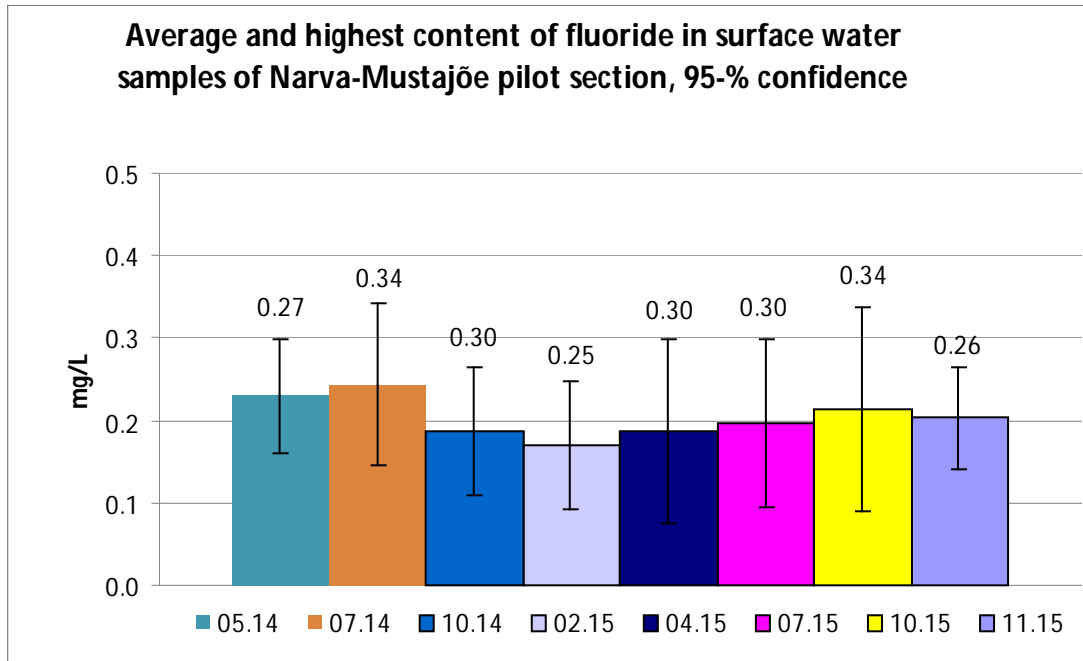


Figure 9. Observed content of fluoride in water samples of Narva-Mustajõe and Simuna-Vaiatu pilot sections (Limit concentration 1.5 mg/L).

As can be seen from Figure 5-Figure 9 the road construction with OSA has mostly affected the content of sulfates in surface water in Narva-Mustajõe. At the same time the content of sulfates in surface water of Simuna-Vaiatu pilot section has decreased. The content of chlorides and values of pH and electric conductivity are similar to natural background levels. Content of fluoride in surface water is well below limit concentration in both pilot sections.

Annual average concentration of sulfates in the surface samples of Narva-Mustajõe and Simuna-Vaiatu pilot sections in 2014 are presented in Figures 10 and 11.

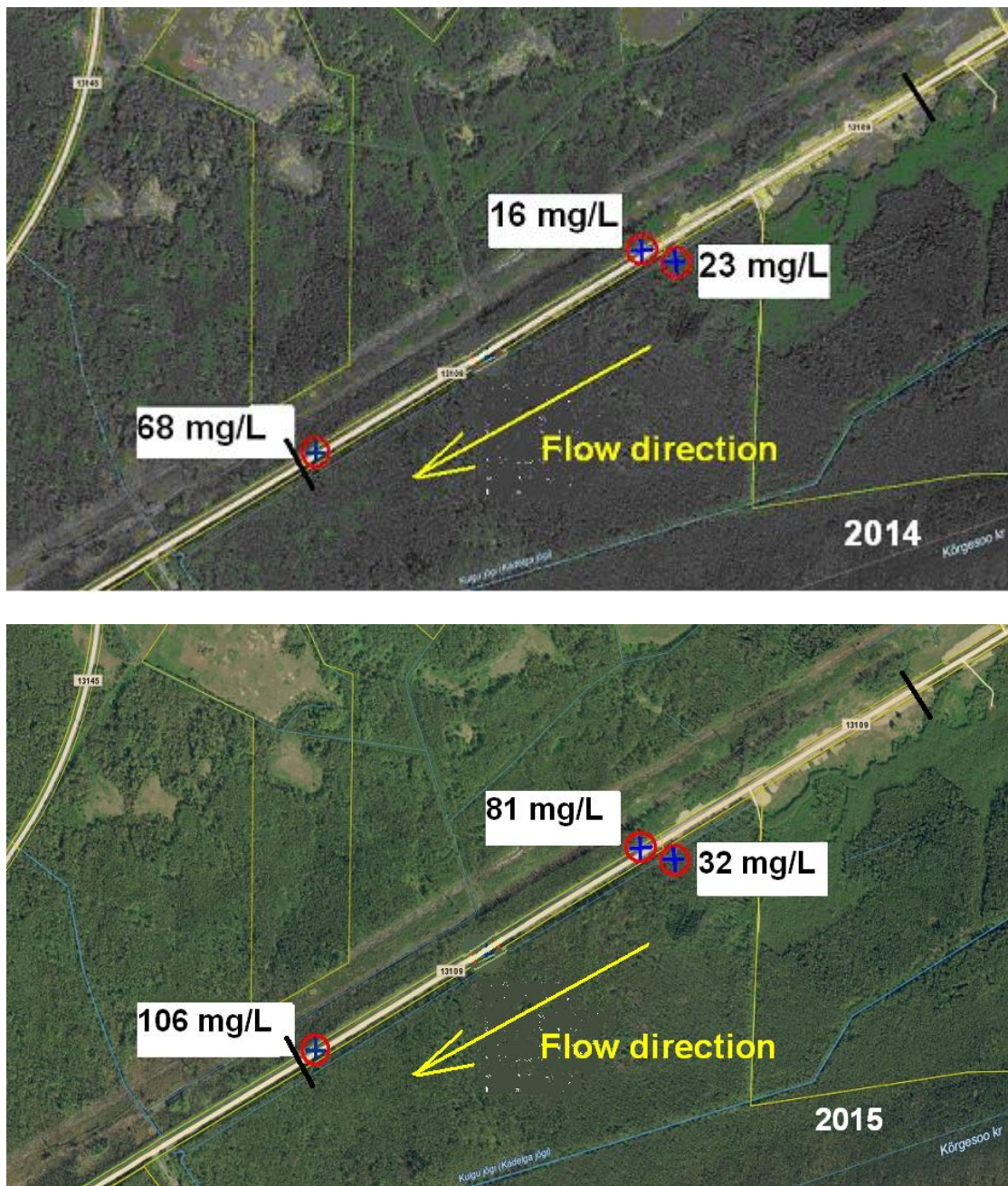


Figure 10 Annual average content of sulfates in surface water sampling points of Narva-Mustajõe pilot section in year 2014 (n=3) and 2015 (n=5).

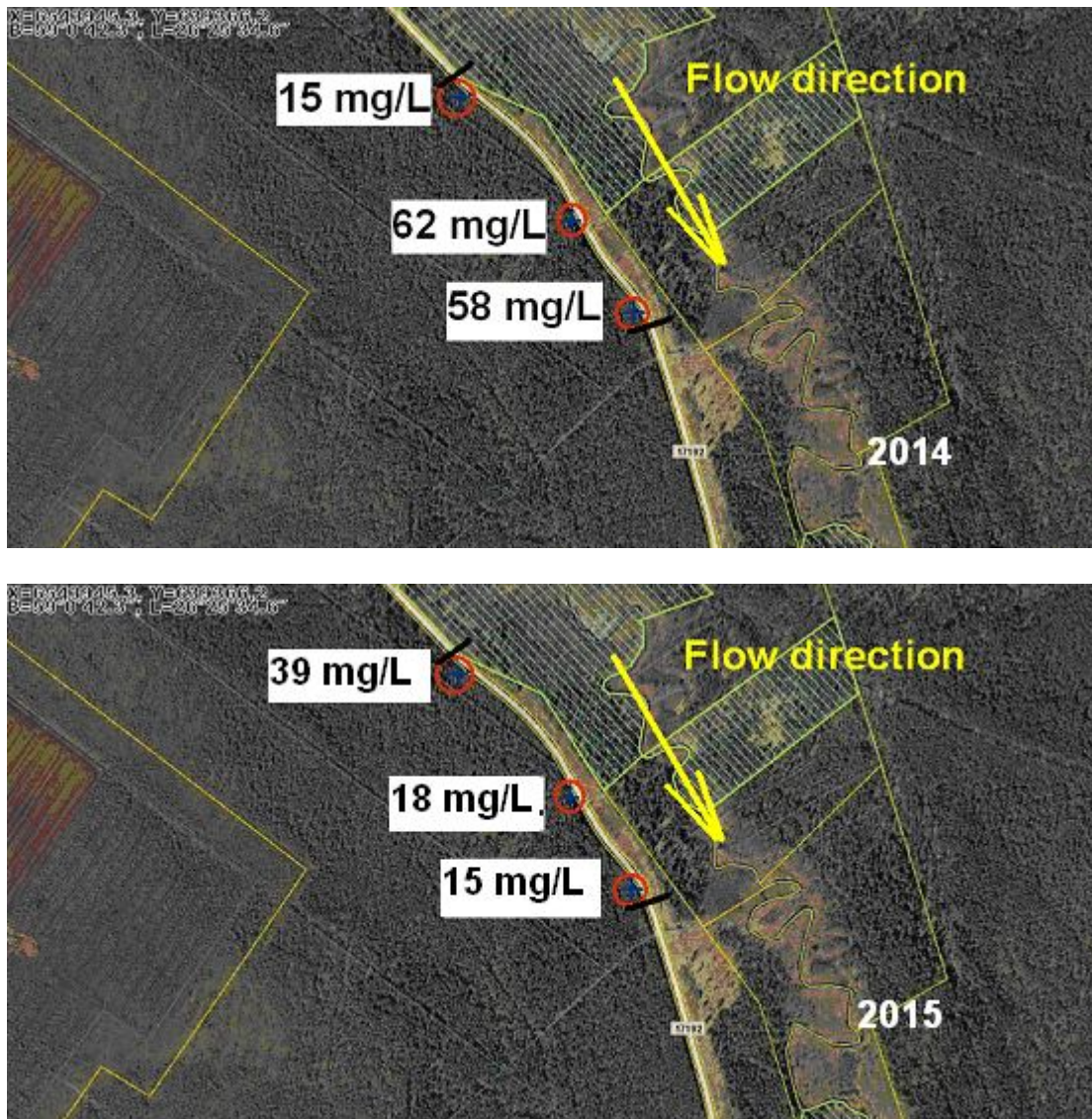


Figure 11 Annual average content of sulfate in surface water sampling points of Simuna-Vaiatu pilot section in year 2014 (n=3) and 2015 (n=4).

As can be seen from figures 8-9 the road construction in pilot sections is affecting the content of sulfates in surface water of Narva-Mustajõe pilot section. In one sample taken downstream of the pilot section (NM2015-1 in 20.10.2015) the content of sulfates was 525 mg/L. The annual average content of sulfates in surface water of Simuna-Vaiatu pilot section is 15-62 mg/L, which not harmful to aqueous environment. Natural level of sulfate in Estonian surface water is 20-50 mg/L and safe content to ecosystem is 80 mg/L according to Institute of Ecology at Tallinn University [10].

Rise in sulfate content in surface water of Simuna-Vaiatu pilot section can be explained by earthworks during road construction in 2014. In 2015 the annual average concentration of sulfates in surface water was lower.



In Narva.Mustajõe section it seems that the road or surrounding environment is affecting the sulfate content in surface water. The concentration of sulfates is rising along the stream and annual average content of sulfates in 2015 is many times higher compared to previous results. It must be noted that according to pH depending leaching test at pH 8 (L/S=10) one kilogram of OSA from 1<sup>st</sup> electrostatic precipitator of CFB boiler can release up to 14 000 mg of sulfate into surrounding aqueous environment [11]. Maximum release of chloride and fluoride at the same conditions is 3 000 and 500 mg/kg, respectively.

### **5.2.3 Trace elements**

Content of following trace elements is presented in following Figure 12-Figure 15 – lead, arsenic, molybdenum and barium. First three metals were monitored in previous programs and are natural constituents in the environment of two pilot sections. In must be noted that in previous monitoring programs the content of trace elements was determined from non-filtrated samples.

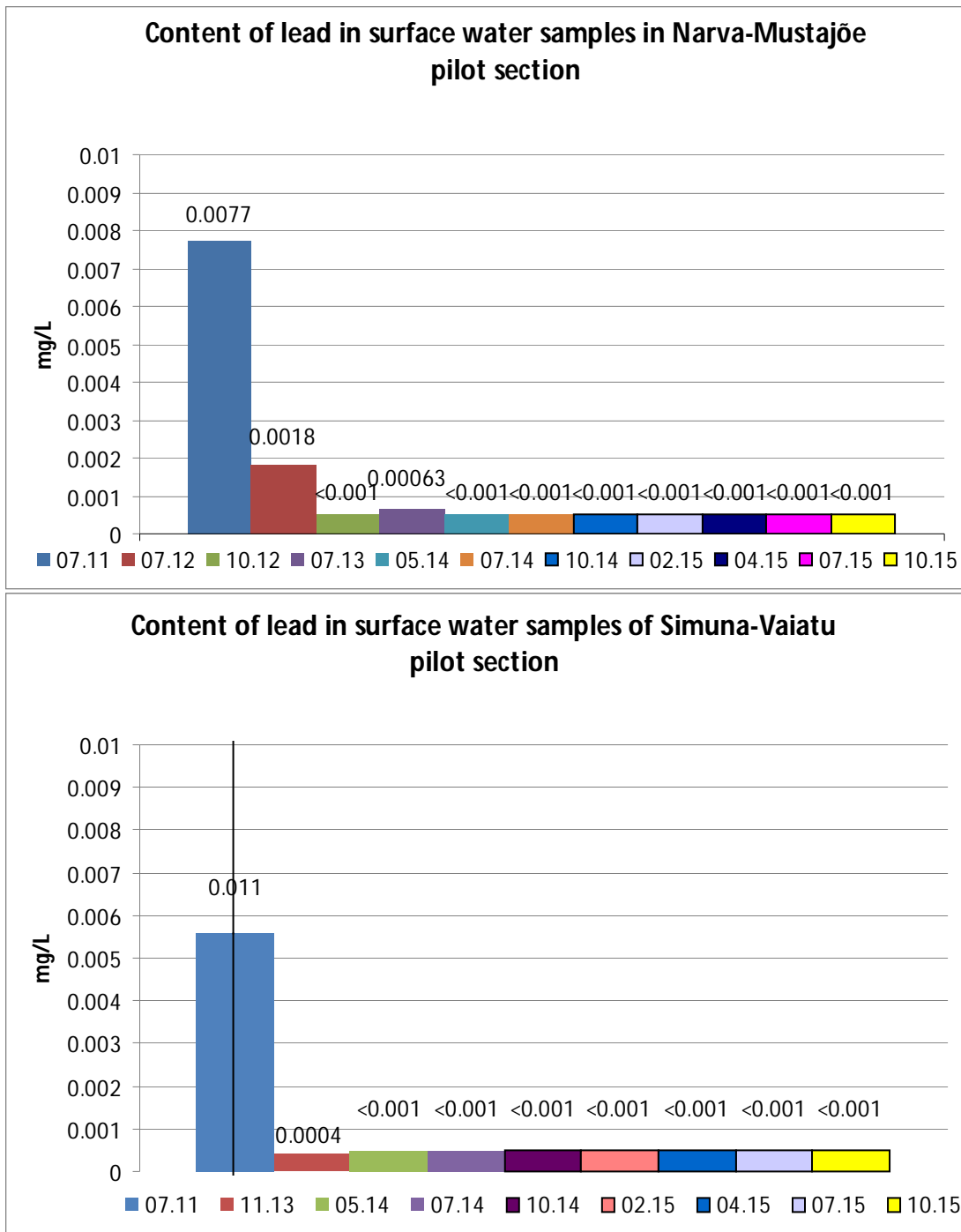


Figure 12 Observed content of lead in water samples of Narva-Mustajõe and Simuna-Vaiatu pilot sections (MAC-ECS for lead in surface water is 0.014 mg/L, 07.11 analysis from non-filtrated sample).

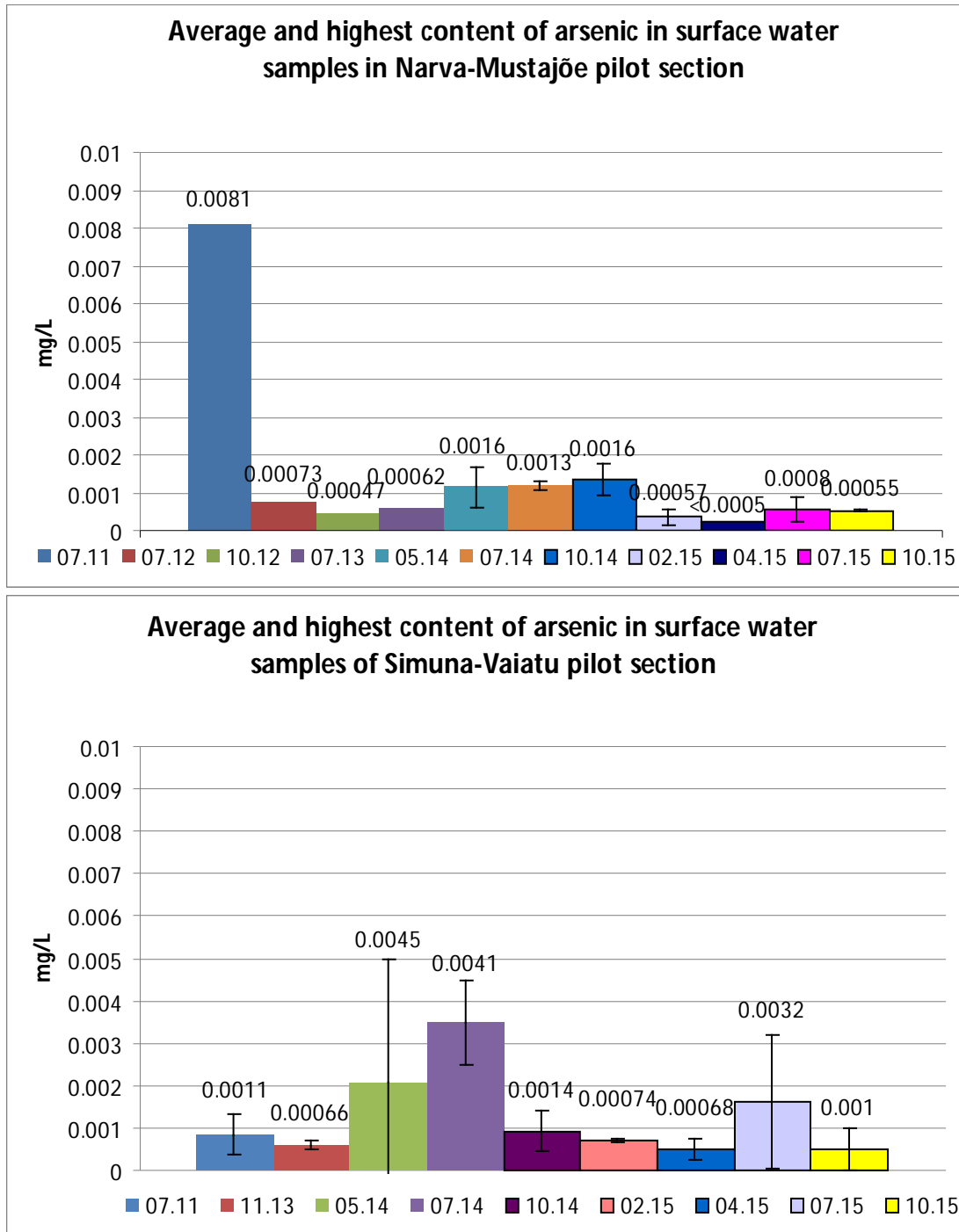


Figure 13 Observed content of arsenic in water samples of Narva-Mustajõe and Simuna-Vaiatu pilot sections (MAC-ECS for arsenic in surface water is 0.01 mg/L, 07.11 analysis from non-filtrated sample).

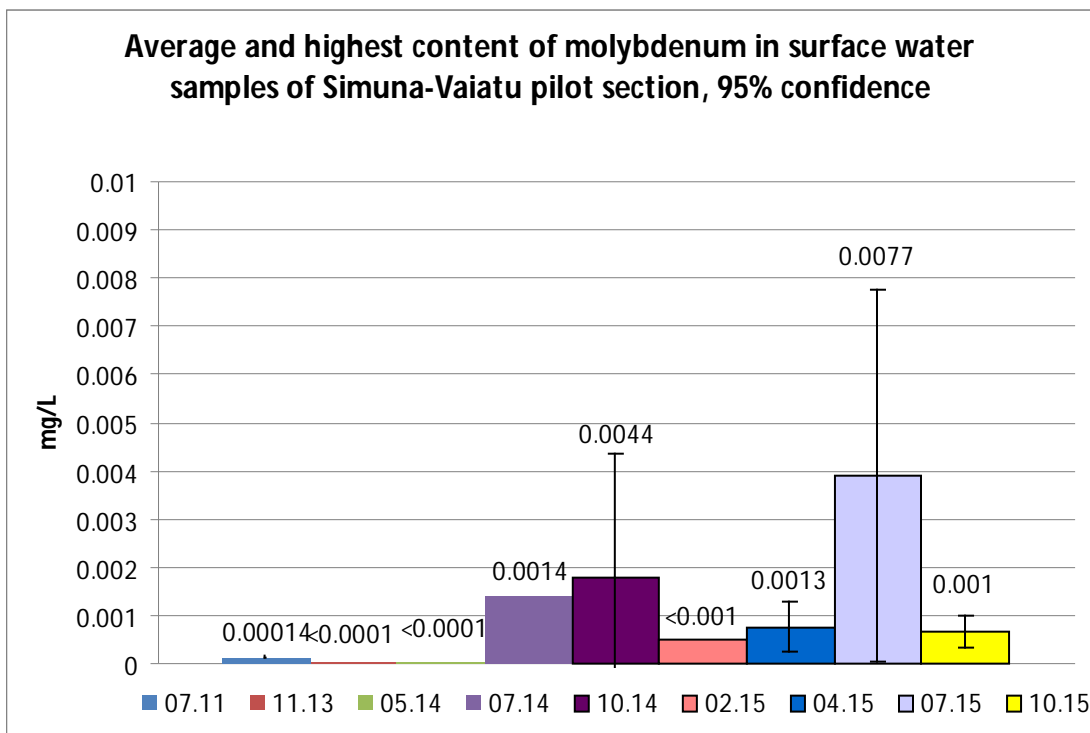
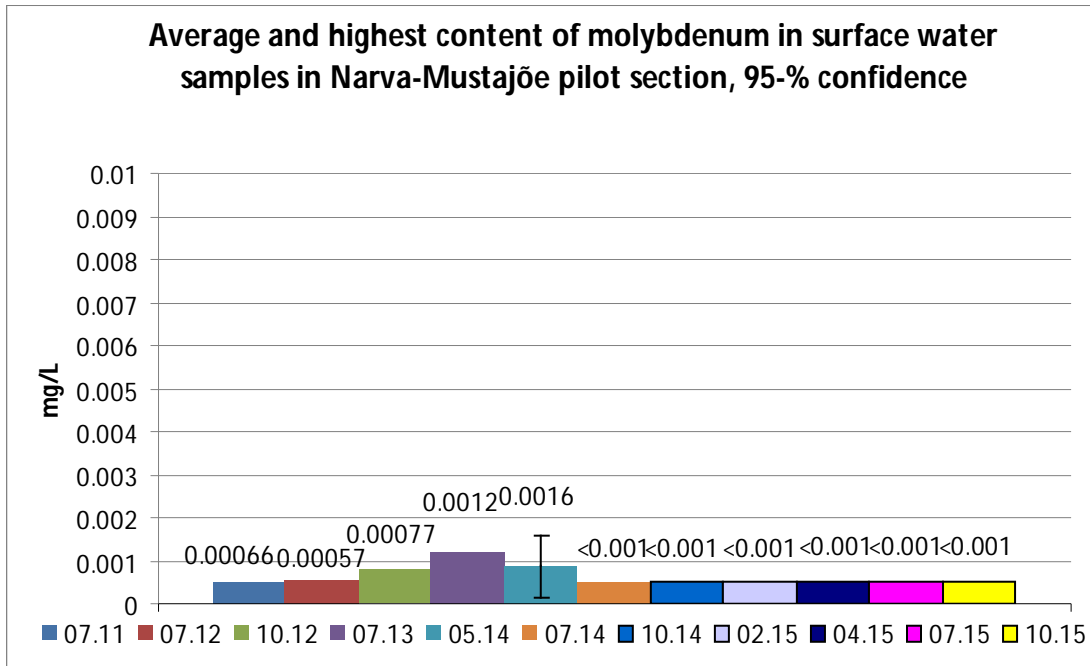


Figure 14 Observed content of molybdenum in water samples of Narva-Mustajõe and Simuna-Vaiatu pilot sections (Limit value in groundwater: 0.07 mg/L).

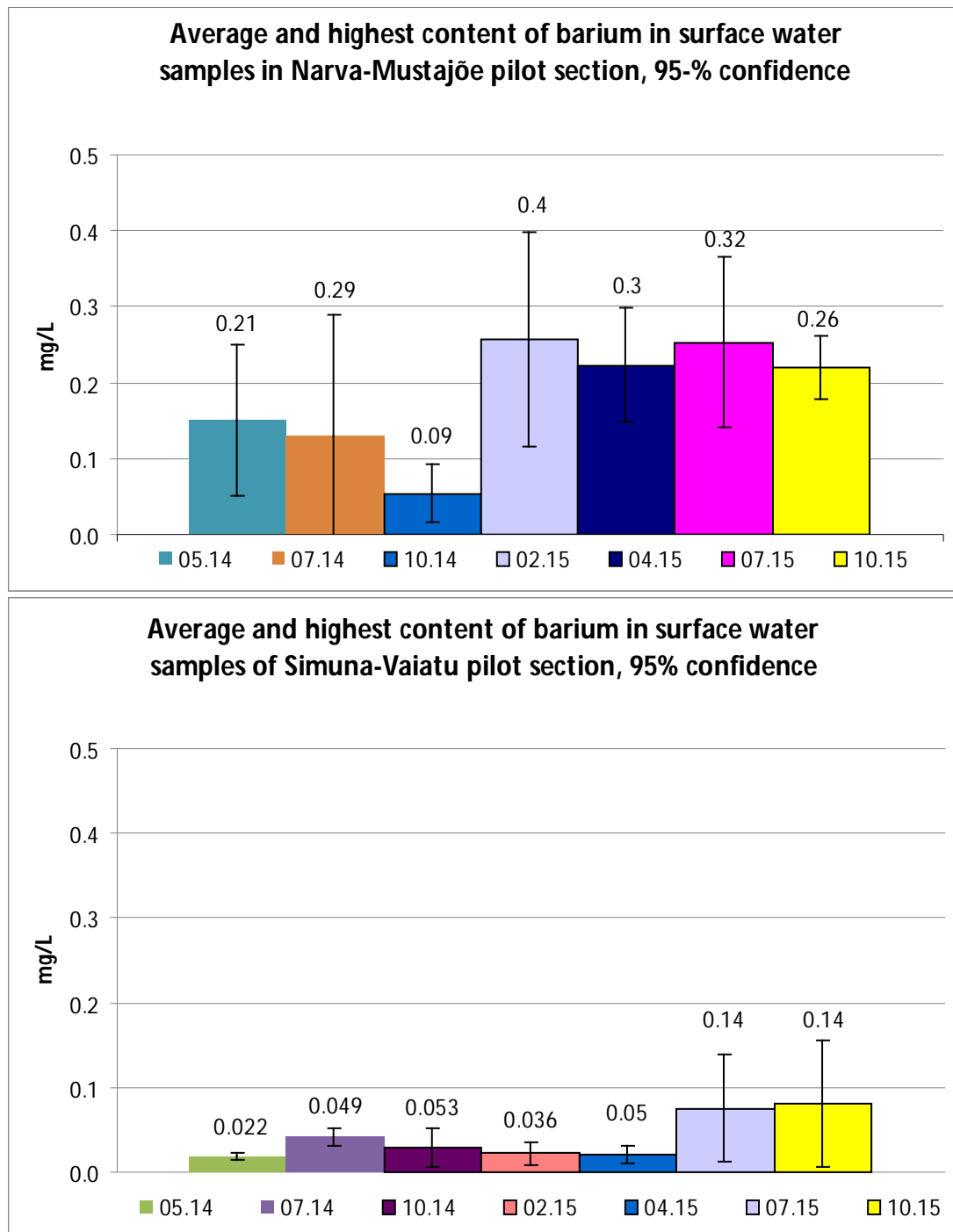


Figure 15 Observed content of barium in water samples of Narva-Mustajõe and Simuna-Vaiatu pilot sections (MAC-ECS for barium in surface water is 0.05 mg/L).

As can be seen from Figure 12-Figure 15 the content of selected trace elements in water samples is below environmental quality standards for lead and arsenic. Also the content of molybdenum is lower than limit value for groundwater. Surprisingly the content of lead has decreased in water samples compared to those taken in 2011 i.e. before road construction. Still, it must be noted that the content of lead and other metals in water samples taken before 2014 are determined from the total sample

content, whilst since 2014 the content of metals was determined from filtrated samples (0.45 µm).

Regarding the content of arsenic and molybdenum different patterns can be seen in Narva-Mustajõe road compared to Simuna-Vaiatu pilot section. In Narva-Mustajõe the content of arsenic has decreased over an order of magnitude compared to first sample taken in 2011. Even if we take the first point as an error no significant changes in content of arsenic can be observed during the monitoring period. There are same fluctuations in the content of molybdenum in water samples of Simuna-Vaiatu pilot section in 2015. But it is well below the national legal limit.

One element, which water-soluble concentration is above national legal limits is barium, especially in Narva-Mustajõe section. Annual average content of trace elements is presented in following figures 13 and 14.

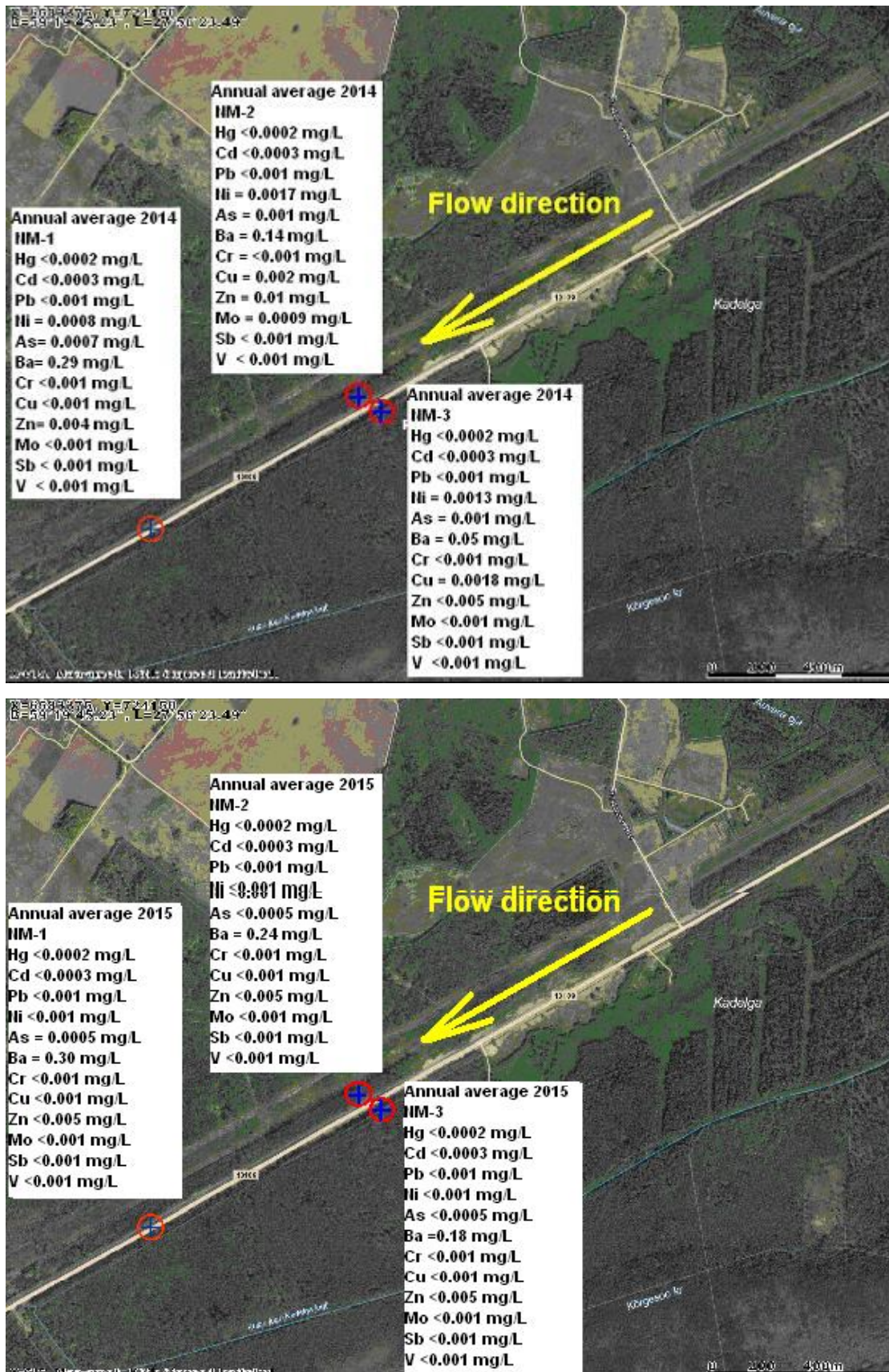


Figure 16 Annual average content of selected trace elements in surface water sampling points of Narva-Mustajõe pilot section in 2014 and 2015.

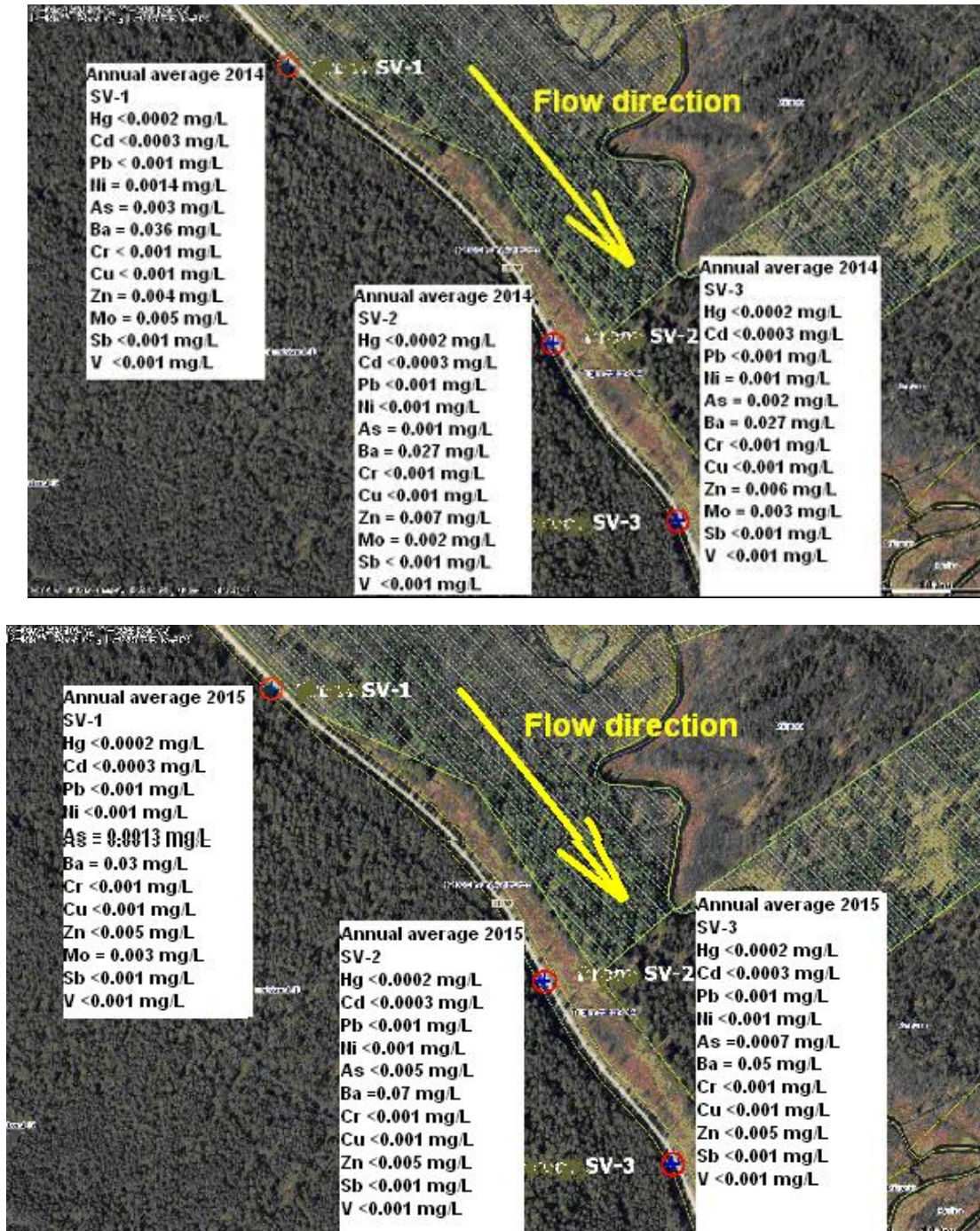


Figure 17. Annual average content of selected trace elements in surface water sampling points of Simuna-Vaiatu pilot section in 2014 and 2015.

As can be seen from figure 16, there is abnormal rise of annual average barium content in surface water samples along the flow direction in Narva-Mustajõe pilot section. Also the content of barium has risen in 2015 in both pilot sections when compared to the results in 2014. At the same time the barium content in soil samples of Narva-Mustajõe has decreased (Table 11 and Figure 18). Barium is a mobile element in the presence of chloride ion and in water comes primarily from natural



sources. The average content of barium in OSA as in mean Estonian –soil is ca 0.3 g/kg and the mobile fraction at pH 8 is 0.3-0.5%.

A surface water sample NM-0 was taken in 21.07.2015 outside the pilot section area in order to establish a natural background level for barium content in surface water in the Narva-Mustajõe road area.

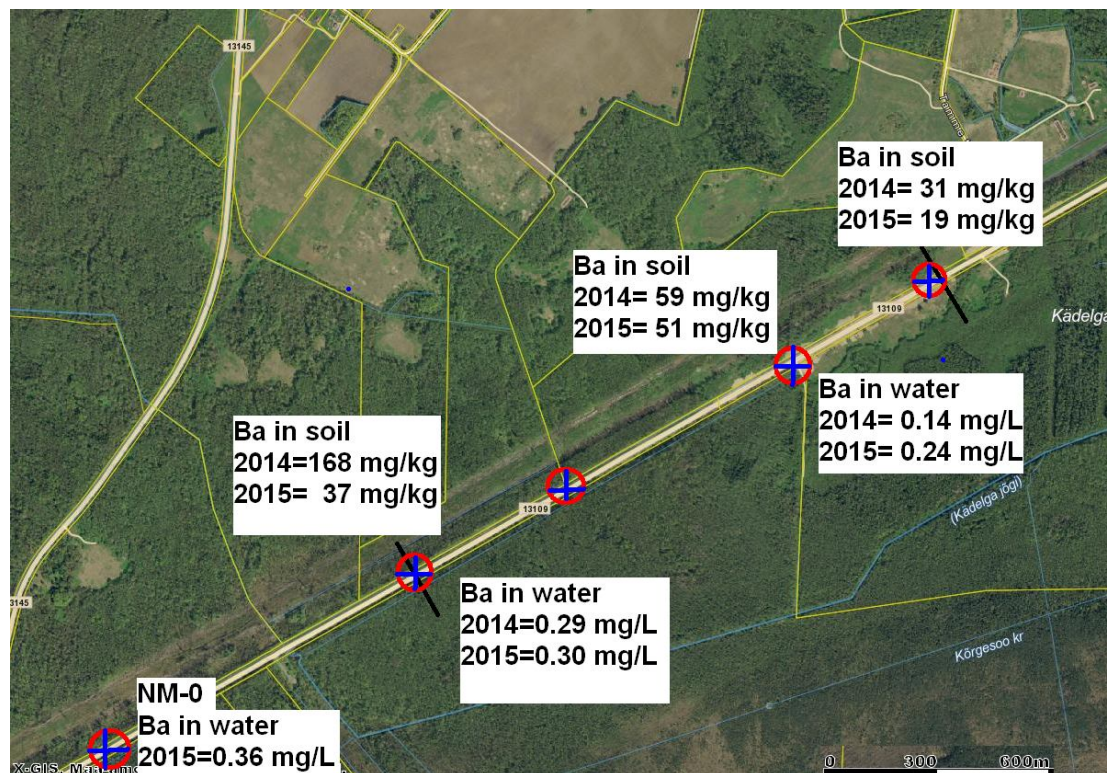


Figure 18. Barium content in soil samples and annual average barium content in surface water samples (except for NM-0).

It turned out that the content of barium in surface water outside the Narva-Mustajõe pilot section area was 0.36 mg/L. This is even higher concentration than maximum annual average concentration of barium in surface (0.29 mg/L in 2014 and 0.30 mg/L in 2015) in the close vicinity of the pilot section area. Latter means that the natural background level of barium in the area is higher than national limits (0.1 mg/L). Ministry of Environment is currently working on amendments in national environmental quality standards [private conversation with Mr. Margus Korsjukov, Senior Officer, Ministry of Environment]. Barium content is **also** naturally high in waters of East-Estonia [7. Conclusions].

The United States Environmental Protection Agency has set the maximum barium level in drinking water 2 mg/L<sup>3</sup>

The Ministry of Environment conducted similar revision for fluoride content during the course of this follow-up monitoring. When the program started the limit concentration for fluoride in surface water was as low as 0.0015 mg/L. By drawing

<sup>3</sup> National Primary Drinking Water Regulations, EPA 816-F-09-004, May 2009

attention to the fact that even natural rainwater contains more fluoride the legal limit was raised up to 1.5 mg/L in 2015<sup>4</sup>.

### 5.3 Groundwater

It is not possible to assess the impact of OSA road construction to groundwater based on the data obtained from the follow-up program. Water samples were taken from the roadside ditches. These ditches act as a discharge for the upper layer of groundwater and infiltration from ditches to groundwater is therefore minimal (Figure 19). No groundwater samples were taken throughout the monitoring program (See appendix 3 Term of Reference, *in Estonian*).

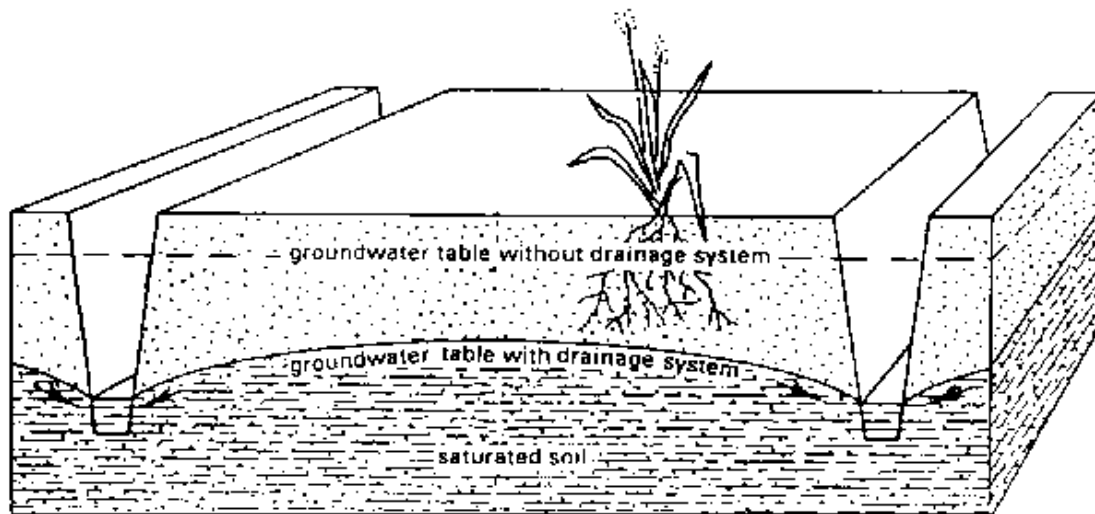


Figure 19. Control of the groundwater table by means of deep open drains [14].

Oil shale ash contains sulfur (up to 4 wt.%, [13]) in many forms and according to leaching tests at neutral pH level one kilogram of OSA releases over 10 grams of sulfates into surrounding aqueous environment [11]. Therefore the content of sulfates in water should be monitored whenever OSA is used as a construction material.

## 6. Data analysis

For deriving hidden information from the collected data a simple bivariate statistical approach was used. Pearson's bivariate correlations between selected parameters in water sampling points of Narva-Mustajõe and Simuna-Vaiatu pilot sections are presented in Table 21-Table 26 with some remarks.

<sup>4</sup> Regulation of the Ministry of Environment, Pinnavee keskkonna kvaliteedi piirväärtused ja nende kohaldamise meetodid ning keskkonna kvaliteedi piirväärtused vee-elustikus, revisions RT I, 18.12.2013, 5 and RT I, 27.03.2015, 20

Table 21. Pearson's bivariate correlations between selected parameters in NM1 during 2014-2015 (blue at significance level  $p < 0.05$ , red at significance level  $p > 0.01$ ).

	<i>Air temp.</i>	<i>Water depth</i>	<i>Water temp.</i>	<i>pH</i>	<i>EC</i>	<i>Cl-</i>	<i>SO42-</i>	<i>F-</i>	<i>Ba</i>
Air temp. oC	1.000								
Water depth, cm	0.589	1.000							
Water temp. oC	<b>0.931</b>	<b>0.800</b>	1.000						
pH	0.700	0.147	0.621	1.000					
EC	-0.717	-0.668	<b>-0.853</b>	-0.444	1.000				
Chlorides	-0.450	-0.405	-0.471	-0.135	0.607	1.000			
Sulfates	-0.451	-0.264	-0.540	-0.242	<b>0.823</b>	0.437	1.000		
Fluoride	0.448	0.120	0.276	0.500	0.173	0.351	0.491	1.000	
Barium	0.299	-0.093	0.172	0.220	-0.173	-0.689	-0.306	-0.288	1.000

Table 22. Pearson's bivariate correlations between selected parameters in NM2 during 2014-2015 (blue at significance level  $p < 0.05$ , red at significance level  $p > 0.01$ ).

	<i>Air temp.</i>	<i>Water depth</i>	<i>Water temp.</i>	<i>pH</i>	<i>EC</i>	<i>Cl-</i>	<i>SO42-</i>	<i>F-</i>	<i>Ba</i>
Air temp. oC	1.000								
Water depth, cm	-0.154	1.000							
Water temp. oC	<b>0.913</b>	-0.196	1.000						
pH	-0.219	0.353	-0.524	1.000					
EC	0.040	-0.500	-0.105	0.052	1.000				
Chlorides	0.378	-0.052	0.196	0.081	<b>0.807</b>	1.000			
Sulfates	-0.369	-0.430	-0.581	0.275	0.608	0.195	1.000		
Fluoride	0.270	-0.592	0.583	<b>-0.887</b>	-0.115	-0.274	-0.301	1.000	
Barium	0.121	-0.593	0.047	-0.009	<b>0.927</b>	0.736	0.367	0.057	1.000

These is significant correlation between barium content and electric conductivity of water in monitoring point NM2.

Table 23. Pearson's bivariate correlations between selected parameters in NM3 during 2014-2015 (blue at significance level  $p < 0.05$ , red at significance level  $p > 0.01$ ).

	<i>Air temp.</i>	<i>Water depth</i>	<i>Water temp.</i>	<i>pH</i>	<i>EC</i>	<i>Cl-</i>	<i>SO42-</i>	<i>F-</i>	<i>Ba</i>
Air temp. oC	1.000								
Water depth, cm	<b>-0.891</b>	1.000							
Water temp. oC	<b>0.910</b>	<b>-0.877</b>	1.000						
pH	0.049	-0.264	0.294	1.000					
EC	-0.616	0.537	-0.525	0.141	1.000				
Chlorides	-0.530	0.496	-0.543	-0.089	<b>0.935</b>	1.000			
Sulfates	-0.233	0.330	-0.476	-0.634	0.489	0.750	1.000		
Fluoride	<b>0.782</b>	-0.604	0.685	-0.426	-0.351	-0.163	0.231	1.000	
Barium	-0.558	0.477	-0.728	0.058	0.513	0.466	0.323	-0.665	1.000

Table 24. Pearson's bivariate correlations between selected parameters in SV1 during 2014-2015 (blue at significance level  $p < 0.05$ , red at significance level  $p > 0.01$ ).

	Air temp.	Water depth	Water temp.	pH	EC	Cl-	SO42-	F-	Ba	Mo
Air temp. oC	1.000									
Water depth, cm	-0.138	1.000								
Water temp. oC	<b>0.896</b>	-0.075	1.000							
pH	0.260	0.344	0.163	1.000						
EC	-0.270	-0.503	-0.249	-0.365	1.000					
Chlorides	-0.209	0.325	0.162	-0.036	0.344	1.000				
Sulfates	-0.168	-0.476	-0.122	-0.337	<b>0.968</b>	0.399	1.000			
Fluoride	0.658	0.109	0.621	0.505	0.134	0.293	0.155	1.000		
Barium	0.031	<b>-0.848</b>	0.010	-0.479	<b>0.812</b>	0.003	0.742	0.119	1.000	
Molybdenum	0.679	-0.374	0.669	0.170	0.343	0.178	0.334	<b>0.856</b>	0.549	1.000

There is a significant correlation between the content of sulfates and electric conductivity of water in monitoring point SV1.

Table 25. Pearson's bivariate correlations between selected parameters in SV2 during 2014-2015 (blue at significance level  $p < 0.05$ , red at significance level  $p > 0.01$ ).

	Air temp.	Water depth	Water temp.	pH	EC	Cl-	SO42-	F-	Ba	Mo
Air temp. oC	1.000									
Water depth, cm	-0.458	1.000								
Water temp. oC	<b>0.942</b>	-0.395	1.000							
pH	<b>0.829</b>	-0.643	0.726	1.000						
EC	0.625	<b>-0.848</b>	0.585	<b>0.835</b>	1.000					
Chlorides	0.327	<b>-0.827</b>	0.347	0.539	<b>0.911</b>	1.000				
Sulfates	0.315	<b>-0.790</b>	0.436	0.450	<b>0.814</b>	<b>0.932</b>	1.000			
Fluoride	<b>0.857</b>	-0.667	<b>0.829</b>	<b>0.955</b>	<b>0.902</b>	0.672	0.626	1.000		
Barium	0.500	<b>-0.841</b>	0.568	0.643	<b>0.929</b>	<b>0.955</b>	<b>0.968</b>	<b>0.791</b>	1.000	
Molybdenum	<b>0.856</b>	-0.474	<b>0.946</b>	<b>0.789</b>	0.702	0.501	0.602	<b>0.899</b>	0.711	1.000

There are significant correlations between content of salts and electric conductivity as well as with barium. Interestingly content of molybdenum is related to fluoride content.

Table 26. Pearson's bivariate correlations between selected parameters in SV3 during 2014-2015 (blue at significance level  $p < 0.05$ , red at significance level  $p > 0.01$ ).

	Air temp.	Water depth	Water temp.	pH	EC	Cl-	SO42-	F-	Ba	Mo
Air temp. oC	1.000									
Water depth, cm	-0.465	1.000								
Water temp. oC	<b>0.976</b>	-0.459	1.000							
pH	0.747	-0.710	<b>0.821</b>	1.000						
EC	<b>0.766</b>	-0.388	0.630	0.327	1.000					
Chlorides	0.743	-0.393	0.618	0.326	<b>0.986</b>	1.000				
Sulfates	<b>0.907</b>	-0.329	<b>0.829</b>	0.545	<b>0.831</b>	<b>0.755</b>	1.000			
Fluoride	<b>0.928</b>	-0.326	<b>0.858</b>	0.578	<b>0.811</b>	0.739	<b>0.997</b>	1.000		
Barium	0.112	-0.191	-0.031	-0.252	0.680	0.730	0.193	0.143	1.000	
Molybdenum	<b>0.882</b>	-0.256	<b>0.826</b>	0.536	0.722	0.631	<b>0.983</b>	<b>0.984</b>	0.040	1.000

Similarly to SV2 the content of salts is related to EC but not to barium. Content of molybdenum correlates significantly with both sulfates and fluoride.

## 7. Conclusions

During the follow-up monitoring program of the OSAMAT pilot road sections following conclusions can be made:

- The content of all selected trace elements in soil samples was below national environmental quality limits during the follow-up monitoring and **the road construction has not affected the soil quality** when compared with the results of previous monitoring campaigns.
- The content of priority hazardous metals, Hg and Cd was below limit of detection (LOQ) in all water samples taken in 2014 and 2015.
- Among priority substances the content of lead was below LOQ in all water samples and traces of nickel were found close to detection limits in both pilot sections. Such a content of nickel in water samples can be considered as a natural background and is not caused by road construction, as can be seen from Figure 17 where the traces of nickel were found in surface water sample upstream of the pilot road section.
- Among hazardous substances following anions and elements were detected: fluoride, arsenic, barium, copper and zinc. **Barium content in Narva-Mustajõe** pilot section was found above national environmental quality standard (0.1 mg/L, Figure 16). At the same time **the natural level** of barium in Estonian surface water ranges between 0.02-0.22 mg/L [12]. E.g. in groundwater close to Narva-Mustajõe section (at Eesti Power Plant) the content of barium has been found at level 4.4-5.2 mg/L [15].
- The natural level of barium in surface and groundwater in East-Estonia is well over national environmental standards and is most probably not affected by the use of OSA in road construction [16]. National environmental quality level of barium is under amendment at the moment.
- Water-soluble content of barium in surface water is mostly affected by the presence of chloride ion. The concentration of chloride ions in surface water is many times higher in Narva-Mustajõe pilot section compared to Simuna-Vaiatu. Latter affects also the content of water-soluble barium chloride concentration in Narva-Mustajõe section. Source for chloride ions could be for example the use of de-icing salt in road maintenance during winter period in Narva-Mustajõe road. In Simuna-Vaiatu the barium is mainly bonded to insoluble salts ( $\text{BaSO}_4$  or  $\text{BaCO}_3$ ).
- **The road construction has not affected the natural level of pH or electric conductivity** of surrounding surface water. Electric conductivity of water is directly connected to dissolved solids or salts. Also **content of chlorides and fluoride is similar to natural background level** or with the observations made before road construction.
- The results in sulfate content in surface water samples in comparison with observations made in pre-construction monitoring leave us believe that **the road construction has raised the sulfate content in surrounding surface water in Narva-Mustajõe but not in Simuna-Vaiatu pilot section.** In 2011 no sulfates were found in surface water samples of both pilot sections, whilst during follow-up monitoring program in 2014 to 2015 elevation in annual average sulfate content in surface water can be observed in Narva-Mustajõe pilot section. Natural level of sulfate in Estonian surface water is 20-50 mg/L.

In one sample taken from surface water in October 2015 from Narva-Mustajõe pilot section the content of sulfates was above 500 mg/L.

- The rise in sulfate content in surface water was observed especially in section where OSA from pulverized firing was used (Figure 1). In that sense the **CFB ash is environmentally friendly alternative** and further developments in utilizing OSA should focus more on ash from CFB boilers.
- It is recommended to follow the content of sulfates in Narva-Mustajõe section in future monitoring campaigns until the content of this compound has stabilized to natural level.

## 8. References

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## **Appendix 1 Accreditations**



## **Appendix 2 Sampling reports and results of analysis**



## Appendix 3 Terms of Reference of the Contract

Lisa nr 1

### Lähteülesande kirjeldus

#### Tööde eesmärk

1. OSAMAT projekti pilootaladel keskkonnaseire teostamine

1.1. OSAMAT projekti põlevkivilendtuha ehitatud Narva-Mustajõe ja Simuna-Vaiatu pilootalade keskkonnaseisundi hindamine ja analüüsimine aastatel 2014 ja 2015 ning põlevkivilendtuha kasutamiseiga seotud võimalike keskkonnamõtjude väljaselgitamine.

1.2. Keskkonnaseire uuringute programm hõlmab vee- ja pinnaseuuringud, mille raames määratakse raskemetallide, kloriidide, fluoriidide ja sulfaatide väljaleostumise intensiivsus. Uuringutes saadud tulemuste põhjal tehakse teaduslik analüüs, mille raames hinnatakse põlevkivilendtuha ehitatud konstruktsiooni keskkonnamõtju ja selle ulatust ning tuha kasutamise keskkonnaohutust teede ehitamisel.

#### 2. Tööde kirjeldus

OSAMAT projekt käiviti 2010. aastal Euroopa Liidu programmi LIFE+ raames. OSAMAT projekti eesmärk on tõestada põlevkivituhaga ehitusmaterjali kasutamise otstarbekust keskkonnakaitsest, tehnilisest ja majanduslikust aspektidest lähtudes. Põlevkivilendtuha katsetati sideainena kahe pilootlõigu väljaehitamisel Narva-Mustajõe ja Simuna-Vaiatu teedel. Narva-Mustajõe pilootlõik asub Ida-Virumaal, Auvere külas, Narva-Mustajõe kõrvalmaanteel nr 13109 kilomeetritel 14,5-16. Pilootlõigu teekatte alus rajati 1630 meetri pikkuselt kihtstabiliseerimise meetodil kolme tuhaliiki kasutades. Kihi paksus oli 25-35 cm.

Simuna-Vaiatu pilootlõik asub Väike-Maarja vallas, Simuna-Vaiatu kõrvalmaanteel nr 17192, kilomeetritel 2,5-5. Simuna-Vaiatu pilootlõik jääb soistele aladele. Projekti käigus katsetati suhteliselt uut tehnoloogiat – turba mass-stabiliseerimist põlevkivilendtuha kasutades. Tehnoloogia järgi segati turvas kohapeal sobiva koguse sideainega. Sideainena kasutati tuha ja tsemendi segu. Stabiliseerimise sügavus oli kuni 4 m.

Aastatel 2011-2013 teostati ülalloodud pilootalade keskkonnaseisundi hindamine ja analüüsimine tuha kasutamise võimalike keskkonnamõtjude hindamiseks.

#### 3. Keskkonnaseire programm

Tabelis 1 on esitatud proovide võtmise programm Narva-Mustajõe ja Simuna-Vaiatul aastateks 2014 ja 2015.

Tabel 1. Proovide võtmise programm Narva-Mustajõe ja Simuna-Vaiatu pilootlõikudel.

	2014. aasta		2015. aasta		
	Proovide ja aruannete arv, tk				
Uuringud	Narva-Mustajõe	Simuna-Vaiatu	Narva-Mustajõe	Simuna-Vaiatu	Proovide ja aruannete arv kokku, tk

Pinnavee proovide võtmine ja analüüsimine	9	9	12	12	42
Pinnaseproovide võtmine ja analüüsimine	3	2	3	2	10
Vahearuande koostamine	1				1
Lõpparuande koostamine			1		1

### 1. Selgitused pinnavee proovide võtmiseks ja analüüsimiseks.

Aastal 2014 võetakse pilootlõikudel 9 pinnaveeproovi: 3 proovi kevadel, 3 proovi suvel ja 3 proovi sügisel.

Aastal 2015 võetakse pilootlõikudel 12 pinnaveeproovi: 3 proovi talvel, 3 proovi kevadel, 3 proovi suvel ja 3 proovi sügisel.

Narva-Mustajõe lõigul võetakse üks proov enne pilootlõigu algust (00+00), teine proov sektsioonis 00+00 kuni 9+50, kolmas sektsioonis 9+50 kuni 16+00.

Simuna-Vaiatu lõigul võetakse üks proov enne pilootlõigu algust (00+00), teine proov sektsiooni keskel, kolmas sektsiooni lõpus.

Võetud pinnaveeproovides analüüsitavad elemendid: Sb, As, Ba, Hg, Cd, Cr, Cu, Pb, Mo, Ni, V, Zn, kloriidid, sulfaadid, fluoriidid, elektrijuhtivus, pH, veetemperatuur.

Iga võetud proovi kohta koostatakse proovivõtmise protokoll, analüüsiakt ning teised uuringuga seotud dokumendid (olemasolul). Dokumendid lisatakse vahe- ja lõpparuannetele.

### 2. Selgitused pinnaseproovide võtmiseks ja analüüsimiseks.

Narva-Mustajõe lõigul võetakse ühel aastal kokku 3 pinnaseproovi: üks proov pilootlõigu sektsioonis 00+00 kuni 9+50, teine sektsioonis 9+50 kuni 16+00 ning kolmas sektsiooni lõpust kolme meetri kaugusel. Proovid võetakse suvel.

Simuna-Vaiatu lõigul võetakse ühel aastal kokku 2 pinnaseproovi: üks proov pilootlõigu sektsiooni keskel ning teine sektsiooni lõpust kolme meetri kaugusel. Proovid võetakse suvel.

Võetud pinnaseproovides analüüsitavad elemendid: Sb, As, Ba, Hg, Cd, Cr, Cu, Pb, Mo, Ni, V, Zn, Se.

Iga võetud proovi kohta koostatakse proovivõtmise protokoll, analüüsiakt ning teised uuringuga seotud dokumendid (olemasolul). Dokumendid lisatakse vahe- ja lõpparuannetele.

### 3. Selgitused vahe – ja lõpparuannete koostamiseks.

Vahearuanne koostatakse 1. detsembriks 2014a. ning lõpparuanne 1. detsembriks 2015a.

Aruanded on sisuliselt teadustöö, mis koosneb sissjuhatusest, sisulisest osast ja kokkuvõttest. Sissejuhatuses peab sisalduma probleemi püstitus, töö eesmärk, uurimisobjektide kirjeldus, keskkonnaseire meetodid.

Sisuline osa peab

- 1) andma ammendava ülevaate tehtud tööst, kuni tulemuste ja järelduste esitamiseni.
- 2) sisaldama probleemi käsitlust kirjanduse põhjal, mille ülesandeks on anda lugejale stardiplatvormi, millelt uuringut alustati.
- 3) käsitlema töös kasutatava metodoloogia valikut ja põhjendust. Kirjeldatakse uurimisobjekti, uurimisvahendeid ja uurimise kulgu etappidena.
- 4) sisaldama töös saadud tulemusi. Tulemuste osas peab olema selline materjal, mis suudab lugejat veenda teostaja poolt kokkuvõttes toodavate lõppjärelduste õigsuses, kuid selles osas lõppjäreldusi veel ei tooda. Töötlemata ja mahukad algandmed pannakse lisadesse.

Kokkuvõte peab sisaldama olulisemaid järeldusi ja ettepanekuid. Kokkuvõte peaks lühidalt sisaldama kõik töö eelnevad osad: sissejuhatuse, probleemi püstituse, ülevaate metodoloogiast, kokkuvõtte tulemustest ja järeldused ning soovitused. Kokkuvõtte peab sisaldama arutelu ka selle kohta, kas püstitatud ülesanne on lahendatud ja mida uut on uurimus andnud. Samuti soovitused, täiendavad ettepanekud edasisteks uurimisteks, sh näiteks töö tulemuste rakendamiseks või uuringute jätkamise suhtes.

VahearuanDES esitatakse aastal 2014 Narva-Mustajõe ja Simuna-Vaiatu pilootlõikudel tehtud uuringute tulemused, analüüs ja esmased kokkuvõtted. Lõpparuandes esitatakse 2014 (vahearuanDE tulemused) ja 2015 Narva-Mustajõe ja Simuna-Vaiatu pilootlõikudel tehtud uuringute tulemused, analüüs ja kogu töö kokkuvõtte. Vahe- ja lõpparuanded koostatakse vastavalt käesoleva peatüki eelmainitud korrale. Vahe- ja lõpparuanded koostatakse inglise keeles. Vahe- ja lõpparuannete koostamiseks antakse teostajale kõik eelnevalt tehtud uuringute tulemused ja teised seire teostamiseks ja kokkuvõtte tegemiseks vajalikud dokumendid.

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