

Research Article

About the United Nations Program "Atmospheric deposition of heavy metals in Europe - estimates based on the analysis of moss biomonitorors"

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Abstract

One of the most important aspects of environmental protection and human health issues is the control of atmospheric air quality. Heavy metals (HM) are among the most dangerous environmental pollutants. In most European countries, the need to study the consequences of their impact on the environment and human health has led to the establishment of national and international programs for the biomonitoring of heavy metal atmospheric deposition. Data on the atmospheric fallout of HMs and other toxic elements are collected based on the analysis of moss biomonitoring, which serve as an analogue of aerosol filters. Under the auspices of the United Nations Commission on Transboundary Transport of Atmospheric Deposition in Europe (UNECE ICP Vegetation), an Atlas of Atmospheric Deposition of Heavy Metals is published every five years. The international team of the sector of neutron activation analysis and applied research of the FLNP JINR have been contributing to these Atlases since 1995. Study of atmospheric deposition of heavy metals and other toxic elements in a number of JINR member and non-member states (Azerbaijan, Albania, Armenia, Belarus, Bulgaria, Greece, Georgia, Kazakhstan, Moldova, Poland, Romania, Slovakia, Western Ukraine, North Macedonia, Serbia and Croatia), including some areas of Central Russia (Moscow, Tula, Tver, Ivanovo, Yaroslavl, Vladimir, Samara, Ryazan' regions, as well as the South Urals and the North Caucasus (Republic of Ichkeria, North Ossetia) made it possible to identify and assess the areas of these pollutions in the studied territories and compare with the levels of similar pollution in Western Europe. The possibility of expanding this program to countries in Asia and the Pacific is currently being discussed.

1. Introduction

Since its adoption in 1979, the Convention on Long-Range Transboundary Air Pollution (CLRTAP) has led to significant improvements in air quality, resulting in reduced acidification, reduced maximum peak levels of ozone and photochemical smog, and reduced concentrations and deposition of heavy metals in the atmosphere.

The LRTAP Convention has also begun to improve levels of atmospheric deposition and nitrogen deposition. Despite this good progress, air pollution in the UNECE region continues to cause serious environmental and health problems, and new problems are emerging.

The moss biomonitor method in combination with

nuclear and related analytical techniques has been regularly used over the past 40 years in Western European countries to study atmospheric deposition of heavy metals (HMs), and over the past 25 years it has become widespread in Eastern European countries. In a number of European countries, the need to study the environmental consequences of heavy metals has led to the creation of national and international programs for biomonitoring of atmospheric deposition of heavy metals. The International Cooperative Programme on Effects of Air Pollution on Natural Vegetation and Crops (ICP Vegetation) was established in 1987, initially with the aim to assess the impacts of air pollutants on crops, but in later years also on (semi-) natural vegetation: "Atmospheric deposition of heavy metals in Europe - assessments based on the analysis of moss biomonitoring" [1].

Increasing the number of ratifications of the Protocol on Heavy Metals, the Protocol on POPs has been identified as a high priority in the new long-term strategy of the Convention. The Convention's future viability depends on the positive and active participation of Parties from all parts of the region, as well as ensuring its broad geographical coverage. Wider ratification and full implementation of air pollution control policies are particularly desirable for countries in Eastern Europe, Caucasus and Central Asia (EECCA: Armenia, Azerbaijan, Belarus, Georgia, Kazakhstan, Kyrgyzstan, Moldova, the Russian Federation, Tajikistan, Turkmenistan, Ukraine and Uzbekistan) and South-Eastern Europe (SEE: (Albania, Bosnia and Herzegovina, Bulgaria, Croatia, Cyprus, Greece, North Macedonia, Montenegro, Romania, Serbia, Slovenia and Turkey). As part of our advocacy efforts in Asia, we also reviewed current knowledge on this issue for the Maley Declaration countries in Southeast Asia (Bangladesh, Bhutan, India, Iran, Maldives, Nepal, Pakistan and Sri Lanka).

At present, the ICP Vegetation comprises an enthusiastic group of scientists from more than 40 countries, including scientists from outside the UNECE region, as the ICP Vegetation stimulates outreach activities to other regions around the world. The aim of the UNECE ICP Vegetation program is to qualitatively and quantitatively characterize the distribution of regional atmospheric deposition in

Europe, highlight the locations of important sources of HM pollution and provide retrospective comparisons through studies that are repeated every five years and published in the European Atlas of Atmospheric Deposition of Heavy Metals and Nitrogen under the auspices of the UN.

Concentrations of heavy metals in mosses correlate well with atmospheric deposition, and the transition to absolute values of heavy metal contents in the air through calibration based on the total amount of precipitation is quite simple [2].

In addition to the determination of HMs, the program also includes the analysis of nitrogen, and since 2010 - persistent organic pollutants (mainly polycyclic aromatic hydrocarbons), since 2015 - radionuclides and, more recently, the analysis of microplastics and cosmic dust. The UNECE is designed to formulate the scientific policy of countries that have signed the UN Convention in the field of studying critical levels of ozone and assessing atmospheric deposition of heavy metals in Europe, using a methodology based on the one-time collection and analysis of moss biomonitoring. Information on the activities of this commission can be found on the website <http://icpvegetation.ceh.ac.uk>. In 1998, 36 countries, including Russia, signed the UN Convention on the Control of Emissions of Heavy Metals into the Atmosphere Using Biomonitoring (Arhus Protocol). The Protocol states that "an approach based on biomonitoring of atmospheric deposition should integrate information to formulate future optimized control strategies, taking into account economic and technological factors." Emissions of cadmium, lead and mercury were determined as they are the most toxic metals. The Joint World Health Organization/Convention Task Force on the Health Aspects of Air Pollution (Health Task Force) conducted a more detailed assessment of the potential health risks from the priority metals – cadmium, lead and mercury – in Europe [3, 4].

Since 2000, the ICP Vegetation has conducted the European moss surveys on heavy metals. It involves the collection of naturally-occurring mosses and determination of their heavy metal concentration at five-year intervals. One of the advantages of using mosses to determine heavy metal concentrations is that it is more cost-effective than long-term monitoring of precipitation, so a higher sampling

Table 1. History of moss biomonitoring surveys in Europe.

Year	Moss collection, number of countries	Coordinators
1968	Moss technique was first proposed	(Åke Röhling and Germund Tyler, Lund University, Sweden)
1975	First nationwide survey in Sweden	Åke Röhling (Lund University, Sweden)
1977	First nationwide survey in Norway	Eiliv Steinnes (Trondheim University, Norway)
1985	First joint Nordic Survey (Denmark, Finland, Norway, Sweden)	Åke Röhling (supported by Nordic Council of Ministers)
1990	Joint Nordic/Baltic survey (adding Iceland, Estonia, Latvia, Lithuania)	Åke Röhling (Lund University, Sweden)
1995	First European survey, 28 countries	Åke Röhling (Sweden) and Eiliv Steinnes (Norway)
2000	Second European survey, 28 countries (UNECE ICP Vegetation)	Alan Buse (ICP Vegetation, Bangor University, United Kingdom)
2005	Third European survey, 28 countries (UNECE ICP Vegetation)	Harry Harmens (ICP Vegetation, Bangor University, United Kingdom)
2010	Fourth European survey, 27 countries (UNECE ICP Vegetation)	Harry Harmens (ICP Vegetation, Bangor University, United Kingdom)
2015	Fifth European survey, 36 countries	Marina Frontasyeva, JINR, Dubna, Russia
2020	Sixth European survey, 39 countries	Marina Frontasyeva, JINR, Dubna, Russia
2025	Seventh European-Asian survey, 45 countries expected	France

density can be achieved.

European surveys have been conducted every five years since 1990, and the latest survey was conducted in 2020/2022 (results not yet published). Mosses were collected from thousands of sites across Europe and their heavy metal, nitrogen and POPs concentrations were determined. Participation from a limited number of EECCA countries mainly concerns the European moss survey for heavy metals, whereas participation in the moss survey in SEE countries is more widespread and also includes nitrogen in a limited number of countries.

The most recent survey of mosses in Europe and beyond was carried out in 2015/2016, involving 36 countries (Table 1), with mosses sampled from approximately 5100 sampling sites, this time involving Armenia, Azerbaijan, Georgia, Kazakhstan, Moldova, Mongolia, Tajikistan and Turkey.

The Report [5] on the latest survey provides data on the concentrations of 12 metals in naturally growing mosses: aluminum (Al), antimony (Sb), arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), mercury (Hg), nickel (Ni), lead (Pb), vanadium (V) and zinc (Zn). Aluminum is a good indicator of mineral particles, mainly windblown soil dust [2, 6], as it is present in high concentrations in the Earth's crust. Antimony is present in very low concentrations

in the earth's crust and is generally considered a good indicator of long-range transport of anthropogenic contaminants [2]. The increase in the production and use of antimony in recent decades has led to the enrichment of Arctic air by more than 50%. Given that the toxicity of antimony is comparable to that of lead, antimony has now replaced lead as a potential toxic trace element in the Arctic atmosphere, which may have broader implications worldwide for ecosystems and human health in the future [7]. Some countries have identified many more additional elements in mosses, including a set of rare earth elements, uranium (U) and thorium (Th), but these are not included in this report. Compared to 1990, the number of participating countries has increased significantly (Table 2).

As an example, Fig. 1 shows a map of arsenic distribution in the study areas. Arsenic concentrations in mosses tend to be low in Northern Europe, Ireland and Germany. High levels of arsenic were observed in southern and eastern countries, with average levels above 1.0 mg/kg reported in Tajikistan, northern Turkey, Kazakhstan, Mongolia, Armenia, northern Greece, Azerbaijan and Romania. In Western Europe, relatively high concentrations of arsenic have been reported in much of France, Rioja (Spain), southern Norway and eastern Iceland. Compared to 2010,

Table 2. Countries (regions) that provided data on moss survey in 2015/2016 [5].

Central Europe	South-Eastern Europe Former USSR Others	Countries of former USSR	Others
Austria ^{N,POPs}	Lithuania ^{N, POPs}	Albania	Armenia
Czech Rep.	Norway ^{POPs}	Bulgaria	Azerbaijan
Denmark (Faroe Islands)	Poland ^N	Greece (samples only from North part of Greece)	Belarus
Estonia ^N	Slovakia ^N	North Macedonia	Georgia
France ^N	Spain	Romania ^N	Kazakhstan
	Roya ^{N, POPs}		
Germany ^{N,CO3}	Sweden ^{N, POPs}	Serbia	Moldova
Island	Switzerland ^{N, POPs}	Slovenia ^N	Russia
Ireland ^{N, CO3}		Turkey (samples only from North part of Turkey)	Tajikistan
Italy, Boltzano ^N			Ukrain

Note: All countries provided data on heavy metals; N and POPs: countries that also reported data on nitrogen and POPs, respectively.

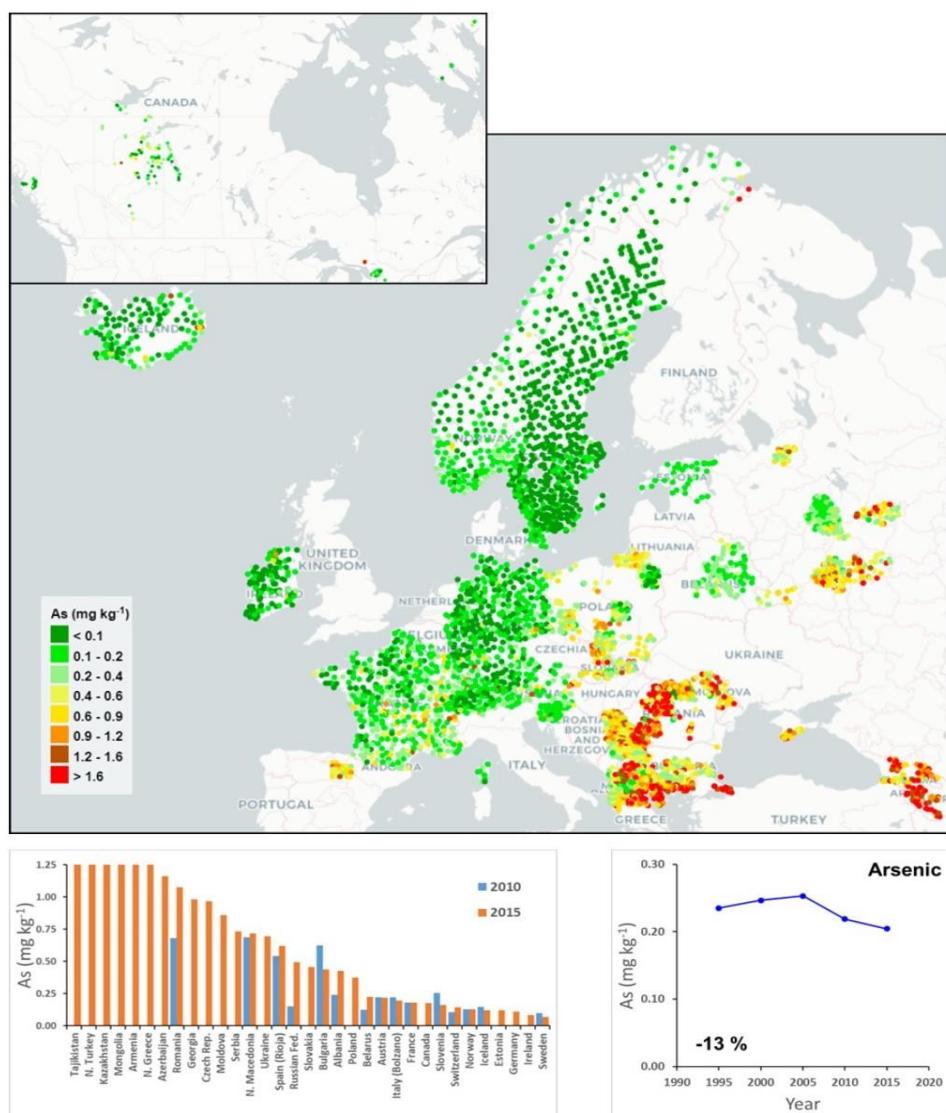


Figure 1. Arsenic concentration in mosses collected in 2015/2016 [5].

average arsenic concentrations increased in Albania, Belarus, Romania, the Russian Federation (where mosses were sampled in additional regions compared to 2010) and Switzerland.

In the past, mosses have been proposed as biomonitoring of POPs: polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs) [8]. For the 2015 study, it was proposed to expand the 2010 pilot study to include additional countries and focus on PAHs, PCBs, polybrominated diphenyl ethers (PBDEs), dioxins and perfluorooctane sulfonic acid and its salts (PFOS), although other POPs could be included if they are of national interest. A recommended list of POPs is available in the 2020 Moss Monitoring Guide [9]. Some countries are interested in data on the distribution of radionuclides (mainly Cs-137) in the study areas [10], as well as in assessing the atmospheric deposition of microplastics [11] and cosmic dust [12].

Similar studies definitely have to be extended to Asia-Pacific, where the concept of the ICP Vegetation program has to be implemented.

As for inhaled airborne particulate matter (PM) which can induce adverse health effects (it is a mixture of many chemical species and varies widely in size, shape and chemical composition, and may contain inorganic ions, metallic compounds, elemental carbon, organic compounds, and compounds from the earth's crust). According to the 2023 World Air Quality Report [XX], the Asia-Pacific region has the top five most polluted countries in the world (<https://www.iqair.com/newsroom/waqr-2023-pr>):

- Bangladesh (79.9 $\mu\text{g}/\text{m}^3$) more than 15 times higher than the WHO PM2.5 annual guideline.
- Pakistan (73.7 $\mu\text{g}/\text{m}^3$) is more than 14 times higher than the WHO PM2.5 annual guideline.
- India (54.4 $\mu\text{g}/\text{m}^3$) is more than 10 times higher than the WHO PM2.5 annual guideline.
- Tajikistan (49.0 $\mu\text{g}/\text{m}^3$) is more than 9 times higher than the WHO PM2.5 annual guideline.
- Burkina Faso (46.6 $\mu\text{g}/\text{m}^3$) is more than 9 times higher than the WHO PM2.5 annual guideline.

Climate conditions and transboundary haze are major factors in Southeast Asia, where PM2.5 concentrations rose in nearly every country.

Although the number of countries and regions monitoring air quality has grown steadily over the

past six years, significant gaps in government regulatory tools remain in many parts of the world. In 2023, air pollution remained a global health catastrophe.

2. Materials and methods

2.1 Moss biomonitor method

Mosses effectively concentrate most heavy metals and other trace elements from air and sediment. Moreover, they do not have a root system and, therefore, the contribution of sources other than atmospheric deposition is limited in most cases. Some types of mosses (*Hylocomium splendens*, *Pleurozium schreberi*, *Hypnum copressiforme*) are distributed over a wide range of temperate climatic zones, and their growing proportion is such that the annual increase can be easily identified. Collection of samples is simple, analysis of mosses is much simpler than sediments, and the exposure period can be accurately determined - usually three years of moss growth are taken for analysis [9].

2.2 Nuclear and related analytical techniques: NAA, AAS and ICP-OES

The use of neutron activation analysis (NAA) to study moss biomonitoring makes it possible to determine up to 45 elements: Ag, Al, As, Au, Ba, Br, Ca, Ce, Cl, Co, Cr, Cs, Dy, Eu, Fe, Hf, Hg, I, In, K, La, Lu, Mg, Mn, Mo, Na, Nd, Ni, Rb, Sb, Sc, Se, Sn, Sm, Sr, Ta, Tb, Th, Ti, V, U, W, Yb, Zn, Zr. The elements Cd, Cu, Hg, and Pb, which are particularly important from an environmental perspective are determined using the method of atomic absorption spectrometry (AAS). The defined set of elements significantly exceeds the number of elements included in the European Atlas (marked in bold). Not all of the above elements are air pollutants; they are determined by multi-element analysis without significant additional costs and can be used as tracers of transboundary air mass transfer. NAA is carried out at the IBR-2 pulsed reactor at FLNP JINR in Dubna using activation by epithermal neutrons along with the full spectrum of neutrons. Measurement of induced gamma activity is carried out using Ge(Li) detectors with a resolution of 2.5-3 keV for the 1332 keV gamma line of ^{60}Co , as well as an HPGe detector with a resolution of 1.9 keV for the 1332 keV gamma line of ^{60}Co .

To process gamma spectra and calculate element

concentrations, a software package developed at FLNP JINR is used [50]. Element contents are calculated by the relative method using certified reference materials Lichen-336 (lichen, IAEA), M1 and M2 (Finnish moss) and Pine Needles (pine needles, NIST).

The determination of Cd, Cu, Hg and Pb is carried out in the chemical analytical laboratory of SNAAPi using atomic absorption spectrometry (AAS iCE3500 Thermo Scientific), and since 2020 using an inductively coupled plasma optical emission spectrometer (ICP-OES, PlasmaQuant PQ 9000 Elite, Analytic, Germany). Mercury is determined by direct measurement by means of direct mercury analyzer DMA-80 evo Milestone.

The error in determining the concentration for most elements is within 5-10%, and only in some cases is 20-25%. The analytical features of both methods are described in the papers cited above [37].

3. Contribution of the joint institute for nuclear research to the UNECE ICP vegetation program

Since 1995, the Sector of Neutron Activation Analysis and Applied Research (SNAAPi) FLNP JINR has been involved in the European program "Atmospheric Heavy Metal Deposition in Europe – Estimations Based on Moss Analysis"). The first contribution to the 1995/1996 Report [13] was the results for the Eastern Carpathians of Romania [14]. In 1999, the project of the NAA and Applied Research Sector "Study of atmospheric deposition of heavy metals in some industrial regions of Russia, Poland, Romania, the Czech Republic, Bulgaria and Slovakia using the moss-biomonitor method and nuclear physics analysis methods and GIS technologies" was included in the problem-thematic plan of the United Institute. Due to the fact that JINR included the countries of Eastern Europe, they were attracted to the UN Air Europe Program and supported their participation in the program with grants from the plenipotentiary representatives of the participating countries in JINR: Poland [15], Bulgaria [16], Czech Republic [17], Slovakia [18] and two grants from the International Atomic Energy Agency (IAEA) - for work in the Southern Urals [19] and Romania [20]. The Sector of the NAA FLNP JINR also carried out pilot studies in

Central Russia (Tula region [21], Tver and Yaroslavl regions [22]) in Western Ukraine [23], in northern Serbia and Bosnia [24], and in 2002 - in Macedonia [25] together with specialists from these countries.

In subsequent years, the number of scientific groups collaborating with FLNP JINR increased significantly. New collaborators included Croatia [26], Greece [27, 28], Albania [29], Italy, the Netherlands, Moldova [30], Mongolia and Tajikistan [31]. Turkey [32], Azerbaijan [33], Armenia [34], Belarus [35] and Georgia [36] also joined the collaboration. The circle of participants within the Russian Federation has expanded: Moscow [37], Vladimir, Leningrad, Samara [38], Ryazan regions, as well as the Southern Urals (Udmurtia) [39] and the North Caucasus (Republic of Ichkeria and North Ossetia [40]).

Additionally, considerable effort was made to involve scientific groups from several universities and high schools in the program to collect biomonitor mosses in their respective regions. The results of these works are reflected in publications, theses and several candidate dissertations. In total, from 2000 to 2010, using instrumental neutron activation analysis (NAA) at the IBR-2 pulsed reactor and atomic absorption spectrometry (AAS) in Dubna, more than 4,000 samples were analyzed. The results were submitted to the UN Commission on Transboundary Transfer of Atmospheric Pollution in Europe for inclusion in the European Atmospheric Deposition Atlas 2000/2001 [41] 2005/2006 [42], 2010/2011 [43] and 2015/2016 [5]. In 2014, by decision of the UN Secretariat, coordination of the Program was transferred from the UK to JINR, which lasted until February 2023. A Data Management System was established on the JINR cloud platform (www.moss.jinr.ru) to collect and analyze information transmitted by program participants to JINR [44]. Similar pilot projects were implemented at SNAAPi FLNP JINR in collaboration with scientists from China [45, 46] and South Korea [47]. During this period, three additional Asian JINR Member States – Mongolia, Vietnam and Kazakhstan – expressed interest in cooperation with the NAA Sector in Dubna. The results of this collaboration are reflected in several joint publications, for example [48, 49]. The latest one-time collection of biomonitor mosses in Europe and not only in Europe in 2020/2021/2022 involved 39 countries, which

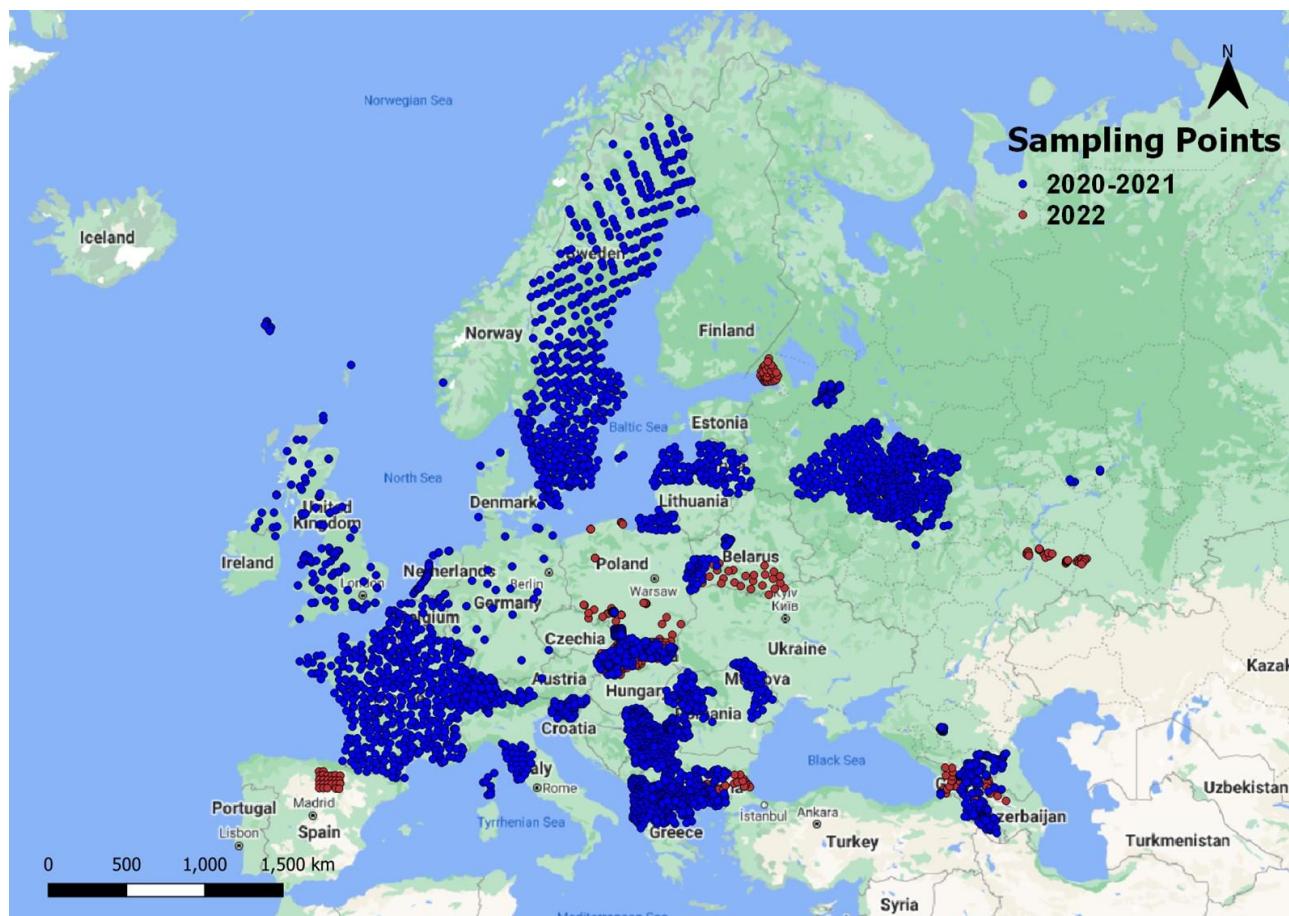


Figure 2. Sampling map of moss biomonitoring in Europe in 2020/2021/2022 (Sampling in 2022 is marked in red).

submitted the results of analysis of more than 4,000 biomonitor moss samples to the www.moss.jinr.ru system. This is what the map of this latest sampling looks like (Fig. 2).

Currently, SNA&AR plans include the dissemination of biomonitoring techniques for the atmospheric fallout of heavy metals, radionuclei, POPs, microplastics and cosmic dust in the countries of Asia and the Pacific region.

4. Conclusions

Existing data on the concentration of heavy metals in mosses from previous simultaneous collections, as well as from the upcoming collection and analysis planned in 2025, are an invaluable source of information for international negotiations and assessment of environmental pollution by heavy metals. The creation of the UN Commission to Study Transboundary Air Pollution was a result of concerns about the level of accumulation of heavy

metals in ecosystems and their impact on the environment and human health.

Data from the one-time collection of moss biomonitoring over large areas (moss survey) allow for the assessment of both spatial and temporal trends of heavy metal concentrations and identification of areas with high levels of atmospheric pollution due to transboundary transport of air pollution. For such an analysis, it is extremely important that sampling be carried out across all of Europe, and now the Asia-Pacific, followed by statistical analysis of moss survey data to understand the factors primarily influencing changes in the concentration of heavy metals and nitrogen in mosses, as well as POPs, radionuclides, microplastics, and cosmic dust.

The use of satellite data will make it possible to predict atmospheric deposition of heavy metals [51, 52]. The implementation of monitoring projects on the territory of European Russia, Siberia and the Far East will allow Russia to contribute to the pan-European monitoring system in Europe and will contribute to

the implementation of this well-tested methodology for studying the areas of pollution by atmospheric deposition of HMs in Russia, especially in areas with significant anthropogenic impact. Regardless of the European Atlas, by Russian specialists with the involvement of modern GIS technologies (geographic information systems) developed in Russia, an Atlas of atmospheric deposition of heavy metals and other toxic elements on the territory of Russia and countries collaborating with JINR can be created.

The expansion of the program to Asia and the countries of the Pacific region will be a new page in the biomonitoring of atmospheric deposition of toxic substances in general.

Disclaimer (artificial intelligence)

Author(s) hereby state that no generative AI tools such as Large Language Models (ChatGPT, Copilot, etc.) and text-to-image generators were utilized in the preparation or editing of this manuscript.

Authors' contributions

It is not applicable because a single author constructed this manuscript.

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Availability of data and materials

All data were published in the cited papers and reports.

Conflicts of interest

The authors declare no conflict of interest

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