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Monitoring atmospheric pollutants in the biosphere reserve Wienerwald by a combined approach of biomonitoring methods and technical measurements

Viktoria Krommer [a], Harald G. Zechmeister [a,b,*], Ingrid Roder [c], Sigrid Scharf [c], Andrea Hanus-Illnar [c]

[a] *Faculty of Life Sciences, University of Vienna, Conservation Biology, Vegetation and Landscape Ecology, Althanstraße 14, 1090 Vienna, Austria*
[b] *ecotox-Austria, Fleschgasse 22, 1130 Vienna, Austria*
[c] *Umweltbundesamt, Spittelauer Lände 5, 1090 Vienna, Austria*

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Abstract

In this study a combined approach of bioindication results correlated with an extensive set of data on air pollution and climate was used to assess the pollution status of the Man and Biosphere Reserve Wienerwald (Austria). Bryophytes served as impact indicators (via the Index of Atmospheric Purity-method IAP) at 30 sites as well as accumulation monitors for airborne trace elements (Al, Pb, V, S, Zn, Fe, Cu, Cr, Ni, Co, Mo, Cd, As, Sb and 16 EPA-PAHs) at 10 sites within the reserve. The results of these bioindication methods were subsequently correlated with further pollution (NO_2, SO_2 and dust) and climate data (precipitation, temperature and humidity).

The findings obtained clearly indicate the following: Bryophyte distribution is solely influenced by the status of air quality, without interference by climatic or site-related factors, which is in contrast to several previous investigations. IAP-values correlated significantly with NO_2 (0.553; $P = 0.004$), SO_2 winter values (0.511; $P = 0.013$) and PM10 (dust) (0.561; $P = 0.013$). The results obtained via chemical analyses revealed a strong correlation with data derived from the IAP methodology. In terms of the overall air quality within the biosphere reserve Wienerwald, the north-eastern part appears to be the most affected one with a most likely pollution contribution emitted by the capital city Vienna, agriculture and neighbouring countries.

Keywords: Air pollution; Index of atmospheric purity; Heavy metals; Mosses; PAHs

1. Introduction

For at least two decades the assessment of environmental pollutants and the menace they pose to the whole ecosystem has been a vital challenge in environmental sciences. Bioindication and biomonitoring have proven to be excellent and cheap ways to observe these impacts of external factors (e.g. Markert et al., 2003).

Bryophytes, due to their morphological and physiological constitution, are indispensable tools within the field of monitoring air pollution: they lack a true root system and a persisting cuticle; thus the uptake of water, nutrients and consequently also toxic substances occurs predominantly via atmospheric deposition (e.g. Bates, 1992). Their reaction to changes in the environment is quick and more direct than those of the majority of vascular plants. In addition, bryophytes show a high degree of sensitivity to certain toxic substances, such as sulphurous or nitrogenous compounds, thus responding by alterations in vitality, abundance and reproduction mode. On the other hand they are strongly resistant to numerous toxic substances and even accumulating them, e.g. heavy metals and a wide

* Corresponding author. Address: Faculty of Life Sciences, University of Vienna, Conservation Biology, Vegetation and Landscape Ecology, Althanstraße 14, 1090 Vienna, Austria. Tel./fax: +43 1 8792994.
E-mail address: harald.zechmeister@univie.ac.at (H.G. Zechmeister).

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range of persistent organic pollutants. Due to their uptake mechanisms of these substances – which permit a correlation between input and concentration, and the possibility of determining the exact period of deposition – they proved as excellent accumulation indicators in many studies (for recent reviews see Zechmeister et al., 2003).

The goal of the Man and Biosphere Program of the UNESCO (2006) is the conservation and sustainable development of selected areas all over the world. These sites outpace confined conservation zones, combining core protected areas with zones where sustainable development is fostered by local dwellers and enterprises. Until now 482 sites – so called 'biosphere reserves' – have been established in 102 countries. The biosphere reserve Wienerwald, due to its exposure to the adjacent capital Vienna, is exposed to a variety of anthropogenic influences. Among these, air pollution plays a major role. Although considerable positive trends could be observed within the last two decades concerning the emission of some atmospheric pollutants, e.g. a remarkable reduction in sulphur dioxide (SO_2) and heavy metals, other compounds such as carbon dioxide (CO_2), total suspended particles (TSP or 'dust'), ozone and PAHs have been increasing or have shown no consistent trend, e.g. nitrogen oxides (NO and NO_2). These facts may have detrimental effects on the total biota of the biosphere reserve (Umweltbundesamt, 2004).

Consequently, the focus of this study was the assessment of the pollution level in the biosphere reserve Wienerwald by four different monitoring networks. Two networks were covered by two different approaches using bryophytes as biomonitors: (1) Assessment of epiphytic bryophytes as reaction bioindicators via the I.A.P. (Index of Atmospheric Purity) approach (LeBlanc and DeSloover, 1970) and (2) Sampling of terrestrial mosses according to international guidelines (Harmens, 2004) and their subsequent chemical analyses for atmospheric heavy metals and the EPA-PAHs. To evaluate the reliability of these analyses, the results of the biomonitoring method were correlated with a set of climatic data (monitoring network three) and data on air pollution obtained by technical measurements (monitoring network four).

2. Material and methods

2.1. Study area

The recently designated biosphere reserve Wienerwald (Biosphärenpark Wienerwald, 2006) spreads to the west and south-west of the Austrian capital city Vienna. About 750.000 people are living in this UNESCO region, which is mainly situated in the province Lower Austria and to a small extent in Vienna, covering a total of 1 054 km^2 (Fig. 1). Geologically and climatically the Wienerwald is located in a transitional zone which can boast of a large-scale broad leave forest unique in biodiversity for Central Europe.

Given numerous roads and motorways with heavy traffic together with the exposure to emission from the adjacent capital Vienna high pollution pressure on the biota of the reserve must be expected.

2.2. Sampling design

2.2.1. Sampling design for mapping epiphytes

The site selection was based on a stratified random sampling design covering the total area of the biosphere reserve Wienerwald and on methodological requirements of the IAP alignment (LeBlanc and DeSloover, 1970). For stratification the following criteria were chosen: (1) consistent distribution of sampling sites within the reserve plus emphasis on exposed border areas and main traffic routes crossing the biosphere reserve; (2) focus on woodland dominated areas (due to the methodology applied); (3) main attention on forest types with high percentage of oak, maple and/or linden. Stratification was performed non-automated, but by using maps and in the field of observations.

A total of 30 sites (five trees each) were investigated (monitoring network one; Fig. 1). Particular emphasis was laid on widely comparable microclimatic conditions at each site (a.o. light, wind and humidity) and bark properties (such as trophic status, pH and humidity). To guarantee the latter the following tree species had been selected (see Zechmeister and Hohenwallner, 2006): *Quercus petraea* (on 23 sites), *Tilia platyphyllos* (on 3 sites), *Acer platanoides* (on 3 sites), *Acer campestre* (on one site). The stems of all sample phorophytes had to be erect, guaranteeing similar deposition patterns and conditions. The determining factor for tree selection within a single site was the availability of a tree meeting the prerequisites described above but not necessarily bryophyte covering.

2.2.2. Sampling design for heavy metal and PAH assessment

Ten samples of terrestrial mosses, each consisting of five to ten sub-samples distributed within an area of 50 m × 50 m, were collected in September 2005 within the overall area following an even distribution of sampling sites and the availability of sufficient material (monitoring network two; Fig. 1). Furthermore we attempted to keep these sampling sites as close as possible to the IAP sites. As biomonitors (Markert et al., 2003), the moss species *Scleropodium purum, Hypnum cupressiforme* and *Abietinella abietina* were used. On the basis of a series of calibration studies showing the uptake efficiency of each moss, heavy metal concentrations determined in these species are fairly comparable (for further reference see Zechmeister et al., 2003). In the laboratory, the moss stems were reduced to the increment of the previous two years, displaying the depositions within this period. The applied method for sampling, sample preparation and analysis is in conformity with the stipulations of the ICP Forests (e.g. Harmens, 2004), which ensures comparability of the data to those of the international programme and ongoing national studies (e.g. Zechmeister et al., submitted for publication). For more confound calculation and drawing of

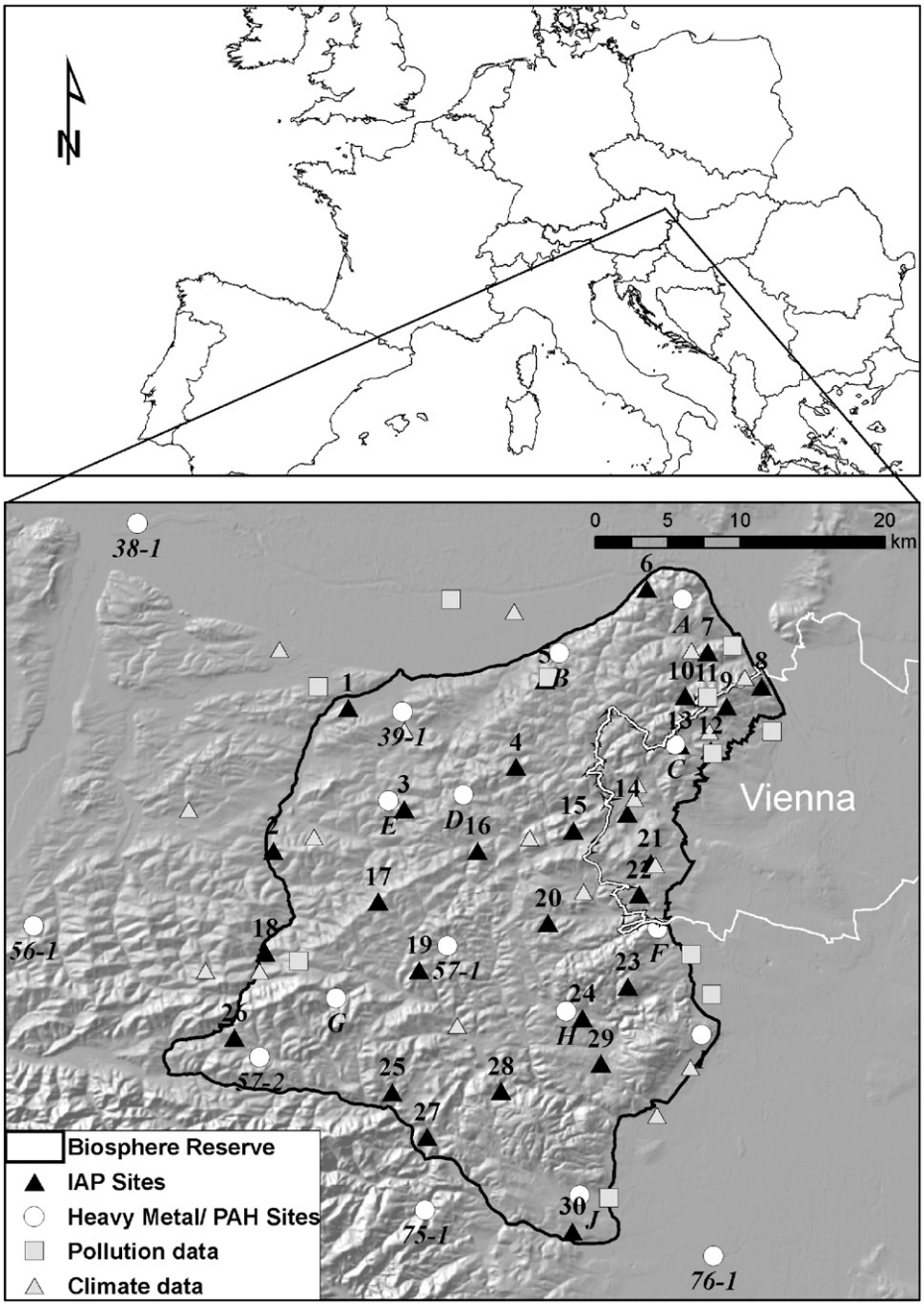

Fig. 1. Map of the biosphere reserve Wienerwald and the location of each monitoring network [IAP sites (monitoring network 1); heavy metal/PAH sites (network 2); pollution data – monitoring sites for the technical measurements of atmospheric pollutants (network 3); climate data – monitoring sites for climatic data (network 4)].

iso-maps data from 17 sampling sites of the international/national program have been included. Three of these sites are located in the Biosphere Reserve; 14 sites were within a distance of 25 km from the biosphere borderline.

2.3. Assessment of epiphytic data according to the index of atmospheric purity (IAP)

Introduced by LeBlanc and DeSloover (1970) to assess overall air pollution, this method has been widely applied in various investigations worldwide (e.g. Sergio, 1987; Palmieri et al., 1997) and further developed by several authors (e.g. Masuch, 1993; Asta and Rolley, 1999; Gombert et al., 2004). It is based on the quantitative and qualitative distribution of epiphytes in the area investigated. In this investigation the following alignment was used:

$$\mathrm{IAP} = \sum_{i=1}^{n} (Q_i \times f_i)$$

n, the number of epiphytic species per site; Q, the resistance factor or ecological index of each species, thus representing

the sensibility of a species against pollutants; *f*, the frequency or coverage score of each species per site. In the present study a frequency scale of three categories was used: (1) rare species and/or species with low degree of coverage; (2) infrequent species and/or species with moderate degree of coverage; (3) frequent species and/or species with high degree of coverage.

IAP-values have been calculated separately for each sampling sites. For the total of all IAP-indices, statistical analyses have been performed including calculation of percentile classes allowing the classification into four air quality groups: group 1 (0–25%), group 2 (25–50%), group 3 (50–75%), group 4 (>75%), with group 1 representing heavy polluted sites.

2.4. Data on atmospheric pollutants measured by technical equipment and climate

Data from 2000 to 2004 on NO_2, SO_2 and dust (as TSP – total suspended particles, respectively as PM10 – particulate matter) obtained by technical measurement were available for 11 sites within or close to the reserve (monitoring network three; Fig. 1). For the years 2000 to 2003 climate data on precipitation and temperature were available for 22 sites and for 7 sites (within those 22) on humidity only (monitoring network four; Fig. 1). Mean annual values for pollution data, temperature and humidity and annual sums for precipitation were calculated for each year, as well as for summer (April to September) and winter (October to March) periods.

As the sites for the assessment of meteorological, air-pollution and IAP data were spatially not identical, a krigging method was used to create an area covering maps for climatic and pollution data, by using an ArcGIS. Respective data for each IAP-sites were derived from these maps by calculating values for each coordinate of an IAP site. Measurement data on pollution were provided by the provincial government of Lower Austria and the municipality of Vienna, climate data were made available by the Central Institute for Meteorology and Geodynamics (ZAMG) and the Federal Ministry of Agriculture, Forestry, Environment and Water Management.

2.5. Chemical analyses

2.5.1. Heavy metals

2.5.1.1. Sample preparation and digestion. Moss samples were dried at <40 °C and ground under liquid nitrogen in a porcelain vessel. For the digestion, aliquots of 1 g (±0.002 g) of ground moss samples were transferred into quartz digestion vessels and 5 ml of nitric acid (s.p. 65%) and 1 ml of perchloric acid (s.p. 70%) were added. In order to avoid loss of volatile elements, absorption vessels, filled with 10 ml of nitric acid, were added to the cooling system. After an overnight reaction time at room temperature, the samples were refluxed in the Kjeldatherm apparatus with an aluminium heating device. Temperature programme: 30 min at 60 °C, 40 min at 200 °C, cooling period 20 min. Once the solutions had cooled down, the contents of the absorption vessels were added to the contents of the digestion vessels, and both the absorption vessels and the digestion vessels and the condenser were rinsed with a further 10 ml of nitric acid.

2.5.1.2. Chemical analysis. The concentrations of Al, Co, Cr, Cu, Fe, Ni, Mo, Pb, V, S, Sb, and Zn were measured by inductively coupled plasma atomic emission spectrometry (ICP-AES). Where necessary, interference was corrected by using the method of multicomponent spectra fitting. Depending on the As concentration, the measurements were performed with the flow injection hydride technique (FIAS) using 0.2% $NaBH_4$ in NaOH (0.05%) as reducing agent or using the FIAS-Furnace coupling technique with an iridium-chloride precoated graphite furnace tube. The measurements of Hg were performed with cold vapour flow injection atomic absorption spectrometry using 0.2% $NaBH_4$ in NaOH (0.05%) as reducing agent. Cd was measured using transversely heated electrothermal AAS (matrix modification with $Mg(NO_3)_2$–$NH_4H_2PO_4$).

2.5.2. PAHs

2.5.2.1. Sample preparation. For the analysis of PAHs in moss samples, soxhlet extraction of 5 g of sample, which had been dried by lyophilisation, was employed by using *n*-hexane as extraction solvent. Afterwards, the soxhlet extract was concentrated to 1 ml in a Turbo Vap (during 35 min, pressure: 1 bar and temperature: 35 °C). A clean-up step was carried out by SPE with special solid phase cartridges for the analysis of PAHs (PAH soil, 1.5 g). The analytes were eluted with a mixture of dichloromethane and petrolether (v:v, 1:4). The extract was once again evaporated to 1 ml and after addition of 10μl of internal standard an aliquot was analysed by GC–MS.

2.5.2.2. Chemical analysis. The following PAHs were analyzed: naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo(*a*)anthracene, chrysene, benzo(*b*)fluoranthene, benzo(*k*)fluoranthene, benzo(*a*)pyrene, indeno(1,2,3-*c*,*d*)pyrene, dibenzo(*a*,*h*)anthracene, benzo(*g*,*h*,*i*)perylene and coronene.

To determine the extraction efficiency of each sample a deuterated PAH cocktail was used as surrogate standard. Fluorantene-C13 was used as internal standard.

The analysis was performed with gas chromatography and mass spectrometry detection (GC–MSD). The ions were detected using selected ion monitoring (SIM).

2.5.3. Quality assurance

2.5.3.1. Heavy metals. Each digestion batch included at least one control sample (moss mixture of 3 different moss samples) or the reference material ''moss 519'', a reference material made available for the European moss survey 1995, and one blank test sample for examination of digestion quality and repeatability standard deviation (mean,

Table 1a
Validity check for chemical methods: Recovery rates and repeatability standard deviation for the determination of heavy metals in reference material "moss 519" (moss 519 – *Pleurozium schreberi*, Finnish Forest Research Institute, MUHOS Research Station) and past data (control charts); for Ba no reference material was available; concentrations of elements are given in μg/g of dry weight; $n = 3$; SD –standard deviation; % SD – percentage of standard deviation

	Al	As	Cd	Co	Cr	Cu	Fe	Hg	Mo	Ni	Pb	S	Sb	V	Zn
Mean	136.3	0.43	0.431	0.99	0.81	70	259	0.082	0.15	16.5	7.3	985	0.16	1.38	37.4
SD	15.5	0.006	0.042	0.08	0.24	5	14	0.021	0.05	0.6	0.7	60	0.02	0.10	1.8
% SD	11.3	1.4	9.8	8.0	30	7.3	5.5	25	37	3.9	9.3	6.1	15.1	6.9	4.9
Target value	*129*	*0.44*	0.44	*0.90*	1.0	68	260	0.062	*0.17*	16.3	8.0	*973*	*0.15*	1.37	36.2
% recovery	*105*	*99.1*	98	*110*	*79*	103	100	133	85	101	91.3	*101*	*105*	100	103

Values in italic are derived not from 'moss 519', but from control charts.

standard deviation and recovery rates for "moss 519"; see Table 1a). Daily instrument performance tests were carried out prior to calibration. After calibration, quality control samples (blank solution, low concentration element mix, high concentration element mix) and spiked moss samples were analysed. External validation of the methods used for moss analysis was performed by participation in inter-laboratory comparisons for moss and lichen samples, arranged by the IAEA (International Atomic Energy Agency).

2.5.3.2. PAHs. For each sample, a surrogate standard of isotope labeled PAHs was added for control of extraction efficiency (see Table 1b). The recovery of each analyte and sample was calculated and used to correct the results. Blank samples were extracted in each sample set to check for contamination during the extraction process. By applying an accredited method, the PAH analysis was monitored continuously by participating successfully in national and international ring examination tests in various matrices.

All analyses were carried out in the laboratory of the Austrian Umweltbundesamt, Vienna.

Table 1b
Validity check for chemical methods: Recovery rates and standard deviation of the corresponding deuterated PAH's in the analysed moss samples

	Recovery rate [%]	SD
Naphthalin-d8	46.8	11.9
Acenaphthylen-d8	61.9	11.0
Acenaphthen-d10	64.6	11.4
Fluoren-d10	74.1	11.4
Phenanthren-d10	86.4	10.6
Anthracen-d10	84.0	9.8
Fluoranthen-d10	94.7	11.6
Pyren-d10	95.6	10.4
Benz(*a*)anthracen-d12	107.3	12.7
Chrysen-d12	94.0	10.4
Benzo(*b*)fluoranthen-d12	108.5	16.1
Benzo(*k*)fluoranthen-d12	100.3	12.8
Benz(*a*)pyren-d12	109.2	15.3
Indeno(1,2,3-*c*,*d*)pyren-d12	105.5	18.8
Dibenz(*a*,*h*)anthracen-d14	114.2	17.2
Benzo(*g*,*h*,*i*)perylen-d12	103.9	15.3

2.6. Statistical analyses

Skewness and kurtosis were calculated to analyse data distribution. ArcGIS (ESRI, version 9) was used for interpolations of the calculated IAP-values – in order to depict belts of varying air pollution burdens – and of site related data (SO_2, NO_2, TSP, PM_{10}, heavy metals, PAHs, precipitation, humidity and temperature) via IDW (inverse distance weighted) algorithm.

Spearman rank correlation was calculated for the correlation between all the measured (m) and calculated (c) variables: IAP-values (m), IAP-classes (m), habitat characteristics (altitude, tree species, habitat type, geology, inclination, exposition, geomorphology, soil humidity and influence by open water; all measured) climatic data (precipitation, humidity, temperature; in total and divided into winter and summer partial amounts; all calculated) and data on air pollution obtained by technical measurements (SO_2, NO_2, TSP, PM_{10}; all calculated).

Multiple regression analysis was calculated to determine the significance of various independent factors influencing element depositions. In a corresponding ANOVA the *F*-ratio, *P*-value and *R*-squared were calculated. Durbin–Watson Statistic was used to test either auto-correlation or serial correlation in the residuals of a least squares regression analysis.

The relationships between IAP-values and environmental variables obtained by technical measurements were calculated via an ANOVA.

2.7. Nomenclature

The nomenclature used is consistent with Grims (1999) for mosses, and Grolle and Long (2000) for liverworts.

3. Results

3.1. Mapping of epiphytical bryophytes

A total of 20 epiphytical bryophyte species (17 mosses and 3 liverworts) was recorded at 30 sites and on 150 trees. Table 2 presents an overview of the taxa in alphabetical order, their calculated *Q*-values and the number of sites

Table 2
Bryophyte species in alphabetical order, their calculated *Q*-values and the number of sites (monitoring network one) they occurred

Species	*Q*-value	∑ sites
Ambystegium serpens	8	1
Bryum laevifolium	6	3
Dicranoweisia cirrata	6.6	5
Dicranum montanum	5	2
Frullania dilatata	4.8	18
Hypnum cupressiforme s.l.	6	4
Leskea polycarpa	6.3	4
Leskeella nervosa	6.5	2
Metzgeria furcata	5.2	10
Orthotrichum diaphanum	4.8	5
Orthotrichum pallens	5	3
Orthotrichum pumilum	8	2
Orthotrichum sp.	6.2	6
Platygyrium repens	7.3	4
Pterigynandrum filiforme var. filiforme	3.5	2
Pylaisia polyantha	4.1	26
Radula complanata	4.8	13
Tortula virescens	7	1
Ulota coarctata	5	4
Ulota crispa var. crispa	4.5	2

they occurred. The most common species in the investigated area of the biosphere reserve Wienerwald are: *Pylaisia polyantha* (on 26 sites), *Frullania dilatata* (18), *Radula complanata* (13) and *Metzgeria furcata* (10). Lichens were not classified at the species level although recorded as single group. The average coverage of lichens was low (8% ± 10%). There was no correlation of the lichen coverage with the IAP values.

Q-values obtained from the present investigation (low *Q*-values indicating higher tolerance towards air pollution, high *Q*-values implicating rather sensitivity) ranges between 3.5 (*Pterigynandrum filiforme*) and 8.0 (*Amblystegium serpens*, *Orthotrichum pumilum*).

3.1.1. Index of Atmospheric Purity (IAP) and technical measurement data

Fig. 2 presents the interpolated IAP-values resulting in zones of different aerial burdens, whereby the north-east appears to be the most affected part of the biosphere reserve.

None of the site-related characteristics (altitude, tree species, habitat type, geology, inclination, exposition, geomorphology, soil humidity and influence by open water) correlated significantly neither with the IAP-values nor the IAP-classes and thus do not seem to exert influence on the two latter.

Table 3 provides mean values of pollution and selected climate data (obtained by data from measurements sites and their subsequent interpolation) for the respective investigation sites. Significant correlations between IAP-values and the following parameters could be detected: NO_2 (0.553; $P = 0.004$), NO_2 summer values (0.575; $P = 0.003$), NO_2winter values (0.552; $P = 0.004$); SO_2 winter values

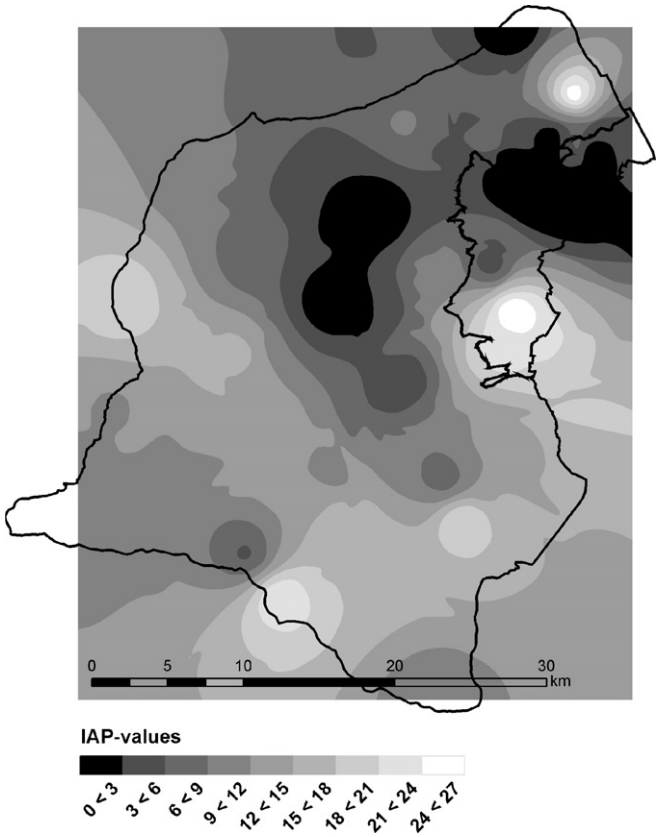

Fig. 2. Zones of varying environmental loads based on interpolation of IAP-values.

(0.511; $P = 0.021$), PM10 (dust) (0.561; $P = 0.013$) and winter temperature (−0.480; $P = 0.008$).

The average IAP-value of 10.6 had its equivalence in 16.8 μg NO_2/m^3, 6.1 μg SO_2/m^3, and 24.2 μg PM_{10}/m^3.

Moreover, winter temperatures correlated significantly with SO_2 winter values (−0.724; $P = 0.000$) and dust (PM10) measurements (0.544; $P = 0.016$). The PCA-Biplot (Fig. 3) supported the relationships mentioned above (cumulative percentage of axis 1 and 2 is 51.8%).

The relationship of seven independent variables to the IAP-value was given in a multiple regression ($F = 3.52$; R^2 adjusted for d.f. 49.4%; $P = 0.031$). Statistically significant contributions to the model were provided by SO_2 ($P = 0.013$), TSP ($P = 0.013$), and humidity ($P = 0.04$).

3.2. Chemical analysis data

3.2.1. QA/QC

Heavy metals: The standard deviations from the analysis of the reference materials were low except for Mo, Hg and Cr. The concentrations of Mo and Cr were also markedly lower than the reference values, and higher for Hg (Table 1a).

As there are only a very few studies available using mosses as monitors of PAHs there is no reference material for PAHs in mosses. However, recovery rates of PAHs were satisfying (Table 1b).

Table 3
IAP-values, IAP-classes, selected site related factors and pollution data: altitude [m a.s.l.]; tree species: 1: Quercus petraea, 2: Tilia platyphyllos, 3: Acer campestre, 4: Acer platanoides; annual precipitation [mm]; annual mean temperature [°C]; mean Winter temperatures [°C]; annual mean values for NO_2 (total, Summer and Winter), SO_2 (total, Summer and Winter), TSP (total suspended particles) and PM10 (particulate matter) [all in $\mu g/m^3$]; ID – corresponding number of monitoring network one

ID	Site_name	Altitude	Tree	IAP-value	IAP-class	Prec.	Temp.	Temp_W	NO_2	NO_2 _S	NO_2_W	SO_2	SO_2_S	SO_2_W	TSP	PM10
1	Eichberg_Plankenberg	241	1	10.2	3	680	11.2	4.8	13	7	15	5	5	5	23	23
2	Hocheichberg_St.Cristophen	336	1	21	4	800	10.3	4.2	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
3	Haberg_Penzing	366	1	7	2	740	10.9	4.8	13	7	17	7	5	6	25	21
4	Troppberg	431	1	0	1	760	10.3	4.2	13	9	17	9	7	7	25	25
5	Tulbinger Kogel	460	1	9.3	3	680	10.3	3.9	9	7	13	13	9	9	25	25
7	Stadtwäldchen_Buchberg	345	2	25.3	4	740	10	3.6	19	13	25	5	5	8	25	29
8	Leopoldsberg	397	2	5.5	2	660	10.9	4.5	19	13	25	5	5	7	25	n.d.
9	Cobenzl	451	2	1.7	1	680	10.6	4.5	15	11	21	5	3	6	23	25
10	Sauberg_Weidlingbach	276	1	0.8	1	720	10.3	4.2	15	11	19	5	3	6	23	25
11	Hermannskogel	499	1	6.5	2	700	10.6	4.2	13	9	17	5	3	6	21	25
12	Schafbergbad	326	4	0.8	1	700	10.6	4.5	19	13	23	5	3	6	25	23
13	Schwarzenbergpark_Neuwaldegg	327	4	2.7	1	720	10.6	4.2	17	13	23	5	3	6	25	23
14	Pulverstampftor_Lainzer Tiergarten	239	1	4.4	1	740	10	3.6	17	13	21	7	5	7	25	25
15	Deutscher Wald_Purkersdorf	333	1	10.6	3	760	10	3.9	17	13	21	7	5	7	27	25
16	Bihaberg_Pressbaum	417	1	0	1	760	10	4.2	15	9	19	7	5	7	27	23
17	Hochstraß	567	1	16	3	800	10.3	4.2	13	9	15	5	5	6	27	21
18	Laaben	411	2	11.7	3	900	9.4	3	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
19	Hainbach_Klausen Leopoldsdorf	456	1	14	3	800	10	3.9	13	11	17	5	5	7	29	21
20	Festenberg_Breitenfurt	399	1	4.4	1	760	10.3	3.9	19	15	23	7	5	8	29	25
21	Lainzer Tor_Lainzer Tiergarten	295	1	27.1	4	700	10.3	4.2	21	17	25	5	3	7	29	25
22	Faßlberg_Mauer	303	4	22.6	4	720	10.3	3.9	23	19	27	5	5	8	31	25
23	Eichberg_Gießhübl	490	1	12.3	3	680	10.3	3.9	23	19	29	5	5	8	31	25
24	Eichkogel_Sparbach	343	1	7.2	2	700	10.3	3.9	21	17	27	n.d.	n.d.	n.d.	n.d.	n.d.
25	Klauswies_Altenmarkt	494	1	5.7	2	900	9.4	3.3	15	11	19	n.d.	n.d.	n.d.	n.d.	n.d.
26	Klammhöhe_St.Corona	612	1	9.9	3	920	9.4	3.3	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
27	Neuhaus	461	1	23.9	4	880	9.4	3.3	17	13	23	n.d.	n.d.	n.d.	n.d.	n.d.
28	Gutental_Meyerling	427	1	17.5	4	780	10	3.6	19	15	25	n.d.	n.d.	n.d.	n.d.	n.d.
29	Siegenfeld_Gaaden	366	1	20.8	4	660	10.3	3.9	21	17	27	n.d.	n.d.	n.d.	n.d.	n.d.
30	Lindenberg_Hirtenberg	340	1	9	2	700	10	3.6	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.

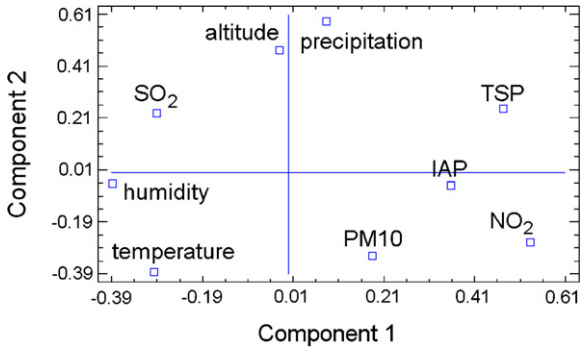

Fig. 3. PCA-component plot of selected variables; "cumulative percentage" of axis 1 and 2 is 51.8%.

3.2.2. Heavy metal concentrations

The average concentrations of 13 analysed sites within the Biosphere Reserve were (± standard deviations): Al 394 ± 257, As 0.15 ± 0.11, Cd 0.25 ± 0.06, Co 0.27 ± 0.13, Cr 0.99 ± 0,53, Cu 8.1 ± 9.1, Fe 503 ± 301, Hg 0.049 ± 0.011, Mo 0.21 ± 0.11, Ni 1.24 ± 0.53, Pb 4.53 ± 1.42, S 1150 ± 176, Sb 0.15 ± 0.06, V 1.14 ± 0.55, Zn 33.2 ± 5.6, N 13433 ± 3502 (for details see Table 4).

Many elements provided evidence of similar spatial deposition patterns with highest concentrations in the east and the north of the Biosphere Reserve. No significant differences between the various sites for all analyzed elements (Friedman Test) could be asserted. Some elements at single sites were highly elevated (e.g. Cu at site J, Cd at site F).

An overall comparison of means of all elements of the all Austrian data (Zechmeister et al., submitted for publication) with those of the Biosphere Reserve showed significantly higher values for the Biosphere data (Wilcoxon matched pairs; $P = 0.001$). However, mean concentrations of single elements were lower in the study area than the Austrian average Pb, Mo and V.

The correlation of heavy metal data with those described in the IAP mapping section represented correlations of Pb with IAP-values (0.510; $P = 0.05$), winter temperature (−0.559; $P = 0.003$) and TSP (0.649; $P = 0.002$); Cu displayed correlations with TSP (0.505; $P = 0.023$).

3.2.3. Polycyclic aromatic hydrocarbons (PAHs)

Concentrations of the analysed PAHs at each site are given in Table 5. Given the fact that no sufficient material could be obtained on one site, data are only available for nine sites. Concentrations at site F were significantly higher than at the other sites (Friedman test; $P = 0.013$). PAH deposition at sites A and B was slightly elevated compared to the others. Concentrations from the Austrian background site were close to the concentrations of most of the sites in the Biosphere Reserve.

PAHs data correlated with TSP (0.736; $P = 0.002$), PM10 (0.782; $P = 0.001$) and winter SO_2 (0.708; $P = 0.003$).

4. Discussion

4.1. Mapping of epiphytical bryophytes

The total number of epiphytical bryophyte species recorded is comparable to other studies with a similar approach (e.g. Sim-Sim et al., 2000; Franzen, 2001; Giordano et al., 2004; Zechmeister and Hohenwallner, 2006). The large majority of them are known as common in Central Europe, although three species can be classified as rare and are meant to be sensitive against air pollution (e.g. *Ulota coarctata*; *Orthotrichum pallens*; *Leskeella nervosa*; Grims, 1999; Sauer, 2000). Species did not show any preference for a certain host-tree species and the various host-trees occurred in a wide range of IAP-values (Table 2). This corrobates other studies (e.g. Zechmeister and Hohenwallner, 2006) that the bark quality of the four host species used are comparable and do not to influence the results.

Comparing the *Q*-values derived from this study and corresponding literature toxi-tolerance-values of Frahm

Table 4
Element concentrations in mosses (μg/g dryweight) at each selected site (monitoring network two) within the biosphere reserve; mv 2005 – mean value of the all Austrian data (derived from Zechmeister et al., submitted for publication; site abbreviations according to Fig. 1)

Site	Al	As	Cd	Co	Cr	Cu	Fe	Hg	Mo	Ni	Pb	S	Sb	V	Zn
39-1	670	0.21	0.22	0.45	2.0	6.0	1000	0.06	0.23	1.8	3.7	1300	0.14	1.6	37
57-1	230	0.11	0.27	0.39	1.0	6.0	450	0.08	0.14	1.8	6.5	1000	0.31	1.0	38
57-2	250	0.07	0.21	0.22	0.73	4.3	420	0.04	0.13	0.75	3.9	880	0.18	0.80	43
A	270	0.11	0.24	0.20	0.67	4.5	320	0.049	0.13	1.3	6.7	1000	0.20	1.1	27
B	260	0.09	0.20	0.21	0.80	4.6	310	0.049	0.31	1.0	3.6	970	0.19	0.77	33
C	370	0.12	0.25	0.20	0.69	5.2	420	0.034	0.13	0.82	2.6	1100	0.11	0.83	35
D	950	0.40	0.34	0.57	1.8	7.0	1100	0.039	0.13	1.6	4.3	1100	0.08	2.5	39
E	140	0.05	0.31	0.13	0.45	4.8	170	0.043	0.13	0.65	2.1	1200	0.07	0.49	33
F	270	0.11	0.35	0.16	0.63	5.3	340	0.051	0.25	0.72	4.9	1100	0.18	1.0	30
G	180	0.07	0.31	0.26	0.66	6.8	240	0.051	0.13	1.6	6.3	1500	0.08	0.86	37
H	840	0.36	0.13	0.36	1.9	5.5	920	0.047	0.21	2.3	4.4	1200	0.11	1.9	24
I	350	0.15	0.20	0.24	0.90	6.1	470	0.056	0.41	1.0	5.4	1200	0.18	1.1	29
J	350	0.13	0.21	0.14	0.75	38	380	0.043	0.44	0.76	4.5	1400	0.12	0.91	27
mv 2005	331	0.14	0.21	0.30	0.81	5.4	420	0.056	1.32	0.24	4.3	965	0.17	1.2	31

Table 5
Concentrations of 16 EPA-PAHs and coronene in mosses at seven investigated sites in monitoring network two (in ng/g dryweight); mv – mean value; SD – standard deviation; background – values of an Austrian background site (according to Zechmeister et al., 2006)

	A	B	C	E	F	G	H	I	J	mv	SD	Background
Naphthaline	2.5	7.9	11	11	1	5.9	13	7.3	2.9	7.3	4.2	6.7
Acenaphthylene	1.4	1.4	0.3	0.3	6.6	0.3	1.0	0.3	0.76	0.6	2.0	0.30
Acenaphthene	3.4	2.3	3.1	5.7	3.0	2.1	2.9	2.3	4.9	3.1	1.2	1.8
Fluorene	4.8	5.2	6.5	6.6	5.6	4.6	5.3	3.8	4.9	4.6	0.9	3.9
Phenanthrene	47	37	33	41	63	36	31	24	30	30.1	11.6	55
Anthracene	2.8	2.1	1.8	1.2	12	1.9	1.7	1.6	1.3	1.6	3.3	1.4
Fluoranthene	55	35	21	13	140	19	15	17	15	16.4	41.0	14
Pyrene	42	25	17	8.5	94	14	12	13	11	12.7	27.2	12
Benzo(*a*)antracene	18	5.7	6.3	2.9	32	5.0	3.7	5.0	3.9	4.4	9.7	1.5
Chrysene	27	14	8.7	5.6	59	9.6	7.8	8.7	7.3	8.4	17.3	4.0
Benzo(*b*)fluoranthene	46	18	9.6	8.3	73	18	12	13	8.8	12.9	22.1	4.3
Benzo(*k*)fluoranthene	18	6.9	5.2	3.6	38	6.1	4.9	5	4.7	5.3	11.3	2.7
Benz(*a*)pyrene	29	11	8.4	5.6	59	9.5	8.0	8.8	7.3	8.4	17.5	3.5
Indeno(1,2,3-*c*,*d*)pyrene	27	15	9.7	8.2	64	13	10	11	9.4	10.8	17.9	2.6
Dibenz(*a*,*h*)anthracene	5.2	1.7	0.5	0.5	9.0	0.5	0.5	0.5	0.5	0.5	3.0	0.81
Benzo(*g*,*h*,*i*)perylene	25	16	8.8	7.1	57	12	9.2	10	9.6	10.3	15.7	3.8
Coronene	6.6	8.4	4.0	3.0	18	5.1	3.1	3.5	2.2	3.5	4.8	3.6
Sum of EPA PAH	*360*	*210*	*150*	*130*	*730*	*160*	*140*	*130*	*120*	137		*120*

(1998), Sauer (2000) and Zechmeister and Hohenwallner (2006) varied remarkably from each other: e.g. according to these studies, *Amblystegium serpens* can be classified as rather insensitive to air pollution, whereas the *Q*-value calculated in this study would reveal sensitivity. The fact that species usually uncommon on a middle European scale appear ubiquitary in the study site and contrariwise frequent bryophytes show less toxitolerance than normally, refers to the exceptional position of the pannonically influenced epiphytical flora of the study site and underlines the importance of calculating sensibility factors of species for each investigation region separately.

4.2. Comparison of IAP-data and data obtained by technical measurements and chemical analysis

For the first time a substantial set of climatic data and data on air pollution obtained by technical measurements could be attributed to IAP-values and classes (see Table 3). There was no correlation between IAP data and site specific or climatic factors. This came a little bit as surprise, as a correlation between these data has been a major concern regarding the IAP-method (e.g. Frahm, 1998; Zechmeister and Hohenwallner, 2006). However, lichen coverage (included in our study) was influenced significantly by tree species, tree vitality, geology, inclination, and vegetation type of the site. Mosses seemed to be more robust to site specific variables than lichens. Bryophyte distribution and the derived IAP-values were obviously mainly influenced by atmospheric pollutants.

The average IAP-value of 10.6 had its equivalence in 16.8 μg NO_2/m^3, 6.1 μg SO_2/m^3, and 24.2 μg PM_{10}/m^3. By interpolation of this total data set of atmospheric pollutants it was possible to depict subsequent belts of varying environmental loads (see Fig. 2, Table 3). To our knowledge up to date no other study so far provided such a dense set of information on a fairly large scale (>1000 km^2) using both, biomonitoring methods and pollution data obtained by technical measurements. The interpolated IAP-values (Fig. 2) indicate a higher pollution impact in the north and northeast of the biosphere reserve Wienerwald. The contribution of the capital, intensive agriculture and neighbouring countries (e.g. Slovakian industries) are likely to represent essential components of these atmospheric pollutions (Umweltbundesamt, 2004).

Comparing results derived from biomonitoring with those of technical measurements revealed a set of interesting correlations: Nitrogen dioxide correlated strongly with the IAP-values indicating the strong influence of NO_2 on bryophyte species distribution. NO_2 pollution in the basin of Vienna and the adjacent parts of the biosphere reserve can manly be attributed to road traffic emissions. Whereas some environmental pollutants (such as e.g. sulphur dioxide, PAHs and heavy metals) indicated overall trends of declining concentrations over the past 10 years, the emissions of nitrogen compounds appeared even to increase, reflecting a continuous growth in transport activities, the general increase of diesel and heavy duty vehicles (Button and Hensher, 2003; Umweltbundesamt, 2004). The same holds for dust (PM10) concentrations – which during the last few years – have been the topic of numerous public discussions concerning increasing public health risks by ultrafine particle emission (Frampton, 2001; Oberdorster and Utell, 2002; World Health Organization, 2005). However, motorways or other roads with heavy traffic did not exert influence on the local level of IAP-values. This corresponds to results obtained by other studies concerning road traffic emissions where contaminations decrease exponentially with distance from the roads and drop to background levels at about 250 m (Viskari et al., 1997; Zechmeister

et al., 2005). Although measurable traffic pollution may be restricted to the vicinity of roads, there is a close suspicion that effects of traffic emissions appear on a larger scale and thus pose threat to the total biota of the biosphere reserve.

In the present study PM10 values revealed a significant correlation with the biomonitoring results, and IAP-values respectively. Especially for PM10 it is known that long range transport from Eastern Europe accounts for high background concentrations in the eastern parts of the study area (Umweltbundesamt, 2004). The negative correlations between winter temperature and SO_2 and dust winter values provide noticeable evidence of the increased emissions by residential heating during the cold season.

Interestingly, only IAP-values and not IAP-classes represented clear correlations with results obtained by technical measurements and moss analyses, which can be attributed to the fuzziness in calculating IAP-classes. This clearly questions the frequent use of IAP-classes. Therefore, we strongly advise the application of IAP-values and their subsequent processing by using GIS applications.

Heavy metal concentrations in mosses were slightly higher at most of the sites than the Austrian average derived from a similar investigation all over Austria. However, based on the results of the QA/QC data of Mo, Cr and Hg have to be considered with caution. The other elements show high congruency with the reference material. Cd and S concentrations in the Biosphere Reserve proved to be remarkably elevated. In comparison with overall Austria, the Biosphere Reserve is situated in an area with generally high loads of deposition, exceeding those of most other parts of Austria (Zechmeister et al., submitted for publication).

Most elements showed analogous spatial deposition patterns in the Biosphere Reserve, with highest concentrations in the north and east, decreasing towards the opposite direction. This underlines that most metals deposited in the Reserve have their main origin in either the adjacent capital or in the intensively used farmlands in the north of the Reserve. These results correspond to the data deduced from the IAP classification. Spatial deposition patterns reveal emission sources outside the Biosphere Reserve for most of the sites. However, some sampling sites might also be strongly influenced by local sources such as local domestic fuel combustion or intensive land-use. Sampling sites close to vineyards showed extremely high concentrations of Cu which is frequently applied as biozide. Cu concentrations at these sites figure among the highest values ever measured in mosses in Austria (Zechmeister et al., submitted for publication).

Long range transport from the refinery in Bratislava might be accounted as source for elevated V, Cr, Pb and Cd concentrations. Cd concentrations are higher in the Biosphere Reserve than in the extended surroundings. Cd has been reported several times as important subject to long range transport and trans-boundary air pollution (e.g. Zechmeister, 1995; Harmens, 2004). Correlation of Cd and NO_2 during summer could point to out a common source (e.g. traffic). Correlations of Pb and Cd with PM10 reveal the importance of these metals regarding their contribution to ultrafine particles.

Highest PAH concentrations were found for phenanthrene, flouranthene, pyrene and benzo(*b*)flouranthene, which is a typical profile for urban surroundings (Orliński, 2002). Recovery rates for PAHs were overall good. Only naphtaline showed lower values, which is probably due to the fact, that naphtaline is mainly occurring in the gaseous phase and therefore can hardly be bound by the moss surface (Zechmeister et al., 2006). PAH concentration did not reveal explicit spatial patterns within the Biosphere Reserve. Concentrations at site F were significantly higher than at the other sites (Table 5). High concentrations of Flouranthen, Pyren and Acenaphtylen provide evidence that the main origin of these depositions is caused by domestic fuel (e.g. Tan et al., 1992) of the adjacent densely populated area. A general correlation of PAHs with winter SO_2 proves the impact and importance of domestic burnings on PAH emissions. However, all sampling sites were more than 300 m from the next house; thus a direct influence from single sources can be excluded. All PAH concentrations were lower than those in comparable studies e.g. from the Warswaw area (Orliński, 2002), from industrial sites in the Czech Republic (Holoubek et al., 2000) or from Hungary (Ötvös et al., 2004). Except for site F and A, PAH concentrations in the investigated Biosphere Reserve were comparable to those given for reference sites in the studies mentioned above.

PAHs data correlated significantly with TSP and PM10 which provide insight to the eminent contribution of PAHs to with ultrafine particles in urban surroundings.

5. Conclusions

The use of bryophytes as biomonitors using the IAP-approach proved to be a perfect tool for the assessment of atmospheric loads, especially NO_2, SO_2 and PM10. Bryophyte distribution was solely influenced by the status of air quality, without interference by climatic or site specific variables. The chemical analysis of various hazardous elements in mosses correlated well with data on overall air pollution obtained by the IAP method. Regarding the overall air quality within the Biosphere Reserve Wienerwald, a fairly strong gradient could be found indicating lowest air quality in the north-east and a strong improvement to the south-west. However, some elements demonstrated diverging patterns which could be attributed to a certain type of land use (e.g. viticulture) or single sources (domestic fuel burnings). Compared to overall Austria the atmospheric burdens are slightly elevated in the east of the Biosphere Reserve decreasing to average values in the western regions. Improvements of this situation are primarily beyond the responsibility of the biosphere management and can only be achieved by national and international efforts.

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