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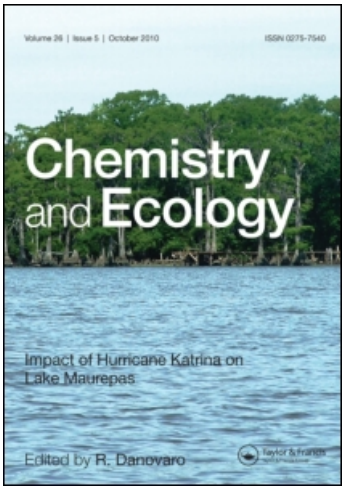

Chemistry and Ecology
Publication details, including instructions for authors and subscription information:
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Elemental concentrations in air, water, and aquatic biota in two rural provinces in northern Vietnam

Annemarie Wagner[a]; Jan B. C. Pettersson[a]; Johan Boman[a]
[a] Department of Chemistry, Atmospheric Science, Göteborg University, Göteborg, Sweden

Online publication date: 21 September 2010

To cite this Article Wagner, Annemarie , Pettersson, Jan B. C. and Boman, Johan(2007) 'Elemental concentrations in air, water, and aquatic biota in two rural provinces in northern Vietnam', Chemistry and Ecology, 23: 1, 63 — 72
To link to this Article: DOI: 10.1080/02757540601083872
URL: http://dx.doi.org/10.1080/02757540601083872

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Chemistry and Ecology
Vol. 23, No. 1, February 2007, 63–72

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Elemental concentrations in air, water, and aquatic biota in two rural provinces in northern Vietnam

ANNEMARIE WAGNER*, JAN B.C. PETTERSSON and JOHAN BOMAN

Department of Chemistry, Atmospheric Science, Göteborg University,
SE-412 96 Göteborg, Sweden

(Received September 2006; in final form 10 October 2006)

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The present study on environmental pollution in northern Vietnam investigates elemental concentrations in fine particulate matter ($PM_{2.5}$), freshwater, and aquatic biota at two sites with differing levels of industrial activities. An Thin is situated 47 km east of Hanoi in the neighbourhood of a coal combustion power plant, whereas the reference site, Duy Minh, is situated in the agricultural province of Ha Nam, 40 km south of Hanoi. Elemental concentrations were analysed using energy-dispersive X-ray fluorescence, total reflection X-ray fluorescence, and graphite furnace atomic absorption spectrometry. All investigated elements in fine particles ($PM_{2.5}$) had significantly higher concentrations in An Thin, thus identifying the air at this site as polluted. In contrast to the aerosol samples, elemental concentrations as well as quantitative differences between the sampling sites were low in freshwater and biota, indicating that the impact of atmospheric deposition was limited.

Keywords: Biomonitors; Dry deposition; Fine particles ($PM_{2.5}$); Freshwater fish (*Clarias fucus*); Freshwater mussel (*Pletholophus swinhoei*); Vietnam

1. Introduction

In recent decades, the Asian continent has experienced an economic development that has resulted in higher demands for energy, mobility, and communications [1]. The consequences of this expansive development include environmental pollution at local and regional levels. In Vietnam, environmental problems are associated with a rapid population growth, resource exploitation, and accelerating industrialization. Many of the larger heavy industries in this country were constructed during an expansive period after the Second World War, when only limited measures were taken to reduce pollutant emissions. The pollution situation has scarcely improved since then and constitutes a growing threat to air and water quality [2].

South East Asia belongs to the regions of the world that are highly sensitive to acid deposition, where negative effects on the terrestrial ecosystem can already occur at sulfur deposition rates exceeding $0.4\,g\,m^{-2}\,yr^{-1}$ [3, 4]. These critical deposition rates are exceeded in some areas in north-eastern Vietnam [5]. Most of the total sulfur deposition in Vietnam occurs on

*Corresponding author. Email: wagnera@chem.gu.se

Chemistry and Ecology
ISSN 0275-7540 print/ISSN 1029-0370 online © 2007 Taylor & Francis
http://www.tandf.co.uk/journals
DOI: 10.1080/02757540601083872

the north coast during September to February, due to transport from China by north-easterly winds. The Chinese sulfur contribution is estimated to make up 40% of all sulfur falling on Vietnam [6, 7].

Sulfur compounds as well as toxic trace metals are mainly found in fine particles with a diameter $<2.5\,\mu m$. These particles result to a large extent from anthropogenic activities such as high-temperature combustion, metal extraction, and refinement processes [8]. A limited number of studies on fine particulate matter ($PM_{2.5}$) in northern Vietnam have been conducted in the capital Hanoi, whereas investigations on $PM_{2.5}$ in rural areas of Vietnam are scarce [9–11]. A recently published investigation on the elemental composition of fine particles conducted at seven rural sites east of Hanoi in the Bac Ninh and Hai Duong provinces revealed concentrations of inorganic species in fine particulate matter of a level comparable with those in the urban area of Hanoi. Coal combustion emissions, fuel oil emissions from river transportation, and, to a minor extent, biomass burning were identified as the main contributors to the inferior local air quality in this rural part of Vietnam [11]. In contrast to the elevated levels of inorganic species found in fine particulate matter, biomonitoring studies using freshwater species collected in northern Vietnam showed generally low levels of trace metals in the organic tissue. This classified water and aquatic biota as unpolluted with regard to trace metal concentrations [12, 13].

The aim of this study is to investigate regional differences in air quality in northern Vietnam. Quantitative differences in the elemental composition of fine particulate matter between two provinces characterized by different levels of industrial activities are studied. These differences are related to elemental concentrations found in freshwater samples and aquatic biota, to assess the impact of air pollutants and deposition processes on aquatic systems and biologic recipients in the investigated area.

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2. Experimental

2.1 *Sampling sites and periods*

All samples, i.e. particulate matter, water, and animals, were collected in two small villages in the northern part of Vietnam, see figure 1. The first site, An Thin, is situated 47 km east of

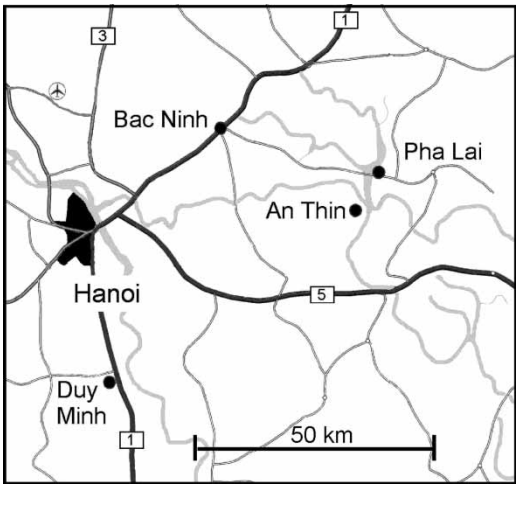

Figure 1. Map of the sampling sites in northern Vietnam.

Hanoi (21.02 N, 105.85 E) and 11 kilometres south-south-west of the Pha Lai coal combustion power plant. Thereby, An Thin is exposed to the emissions of the power plant when the wind direction is north or north-east. The second site, Duy Minh in Ha Nam province, is situated approximately 40 km south of Hanoi. This province was selected as a reference sampling site, since it is mainly agricultural without any industrial areas or larger factories.

The aerosol samples—fine particles with a diameter $<2.5\,\mu m$—were collected during a measurement campaign in November to December 2000. The campaign comprised 15 measurement periods of approx. 48 h each at both sites. The sample collectors were located on the roofs of official buildings at a height of approximately 10 m above ground level. At the same places, rain samples were collected in acid-cleaned polyethylene containers. The rain samples were collected at the same intervals as the particle samples.

The biomonitoring samples consisted of 10 catfish (*Clarias fucus*) and 10 mussel (*Pletholophus swinhoei*) samples collected at each site. All animals were of approximately the same age and, within the same species, of same size or weight [12, 13]. Water samples were taken at each site together with the sampling of biologic material during the second week of March 2001. At the reference site Duy Minh, the freshwater samples were taken in a lake approximately 500 m west of the national road 1A running from north to south, and in An Thin, they were collected from a lake situated at a distance of 1 km from the village [12].

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2.2 *Sample treatment and analytical procedures*

The fine particle samples were collected with Higgins-Dewell cyclones (Casella Ltd, Nottingham, UK). Teflon filters with a 21-mm effective diameter and 3-μm pore size were used as collection substrate. The air flow was generated by a vacuum pump and adjusted to $3.1\,l\,min^{-1}$, in order to obtain a cut-off of 2.5-μm aerodynamic diameter. The flow rate was limited by critical orifices.

The elemental analysis of the particle, rain, and surface water samples was mainly done with an Energy Dispersive X-Ray Fluorescence (EDXRF) spectrometer. Some trace metals in the freshwater samples, however, have been analysed by a graphite-furnace atomic absorption spectrometer (GF-AAS) due to their low concentrations. The EDXRF spectrometer used in this study is based on a three-axial geometry, which results in lower background radiation compared with a conventional setup without a secondary target [11, 14]. For the spectral analysis and quantitative calculations, the AXIL/QXAS package was used [15]. The biological samples were dissected in Hanoi at the National Center for Natural Science and Technology, Institute of Biotechnology. From the catfish, muscle and liver tissue were chosen for analysis, whereas from the mussel samples, the whole soft tissue was used. For the elemental analysis of the biologic samples, a total-reflection X-ray fluorescence spectrometer (TXRF) and a GF-AAS were used. The different organic tissues, i.e. fish muscle, fish liver, and mussel soft tissue, were treated and analysed as described elsewhere [12, 13].

2.3 *Quality assurance*

The accuracy of the cyclones used for the collection of fine particles was tested in a separate study. The study showed a good reproducibility between the total masses of the fine and coarse particle fraction measured by the cyclone and an impactor, respectively, with a high correlation of $r^2 = 0.86$ (number of samples: 8) [16].

The EDXRF detection limits are described elsewhere [17]. The accuracy of the EDXRF method was tested with standard reference material 1577a bovine liver. The results of this test, which are in good agreement with the certified values, are presented in table 1.

Table 1. Comparison of certified elemental concentrations ($\mu g\,g^{-1}$) in SRM 1577a bovine liver and the results obtained with the EDXRF used in this study.

SRM 1577a	S	K	Mn	Fe	Cu	Zn	Rb
Certified values	7800 ± 100	9960 ± 70	9.9 ± 0.8	194 ± 20	158 ± 7	123 ± 8	12.5 ± 0.1
Values analysed by EDXRF	7000 ± 800	9800 ± 850	11.2 ± 2.3	211 ± 13	168 ± 9.4	135 ± 8	14.0 ± 1.2

Note: The measured values are given as the mean concentrations of eight measurements ± 1 S.D.

For the organic samples, the accuracy of the analyses was tested using standard reference material CS ZC 78005 mussel (China National Analysis Center for Iron and Steel, Beijing, China). The standard reference material was treated in the same way as the samples with respect to preparative procedures including digestion, drying, and analysis. The results were in good agreement with the certified values and are described elsewhere, together with the calculated detection limits for TXRF and GF-AAS [12].

3. Results and discussion

3.1 *Fine particle ($PM_{2.5}$) samples*

The mean elemental concentrations and standard deviations of $PM_{2.5}$ samples concomitantly collected in An Thin and Duy Minh during November and December 2000 are presented in figure 2. All elemental concentrations were significantly higher in An Thin, where elemental concentrations clearly exceeded those typically found in rural and semi-rural European

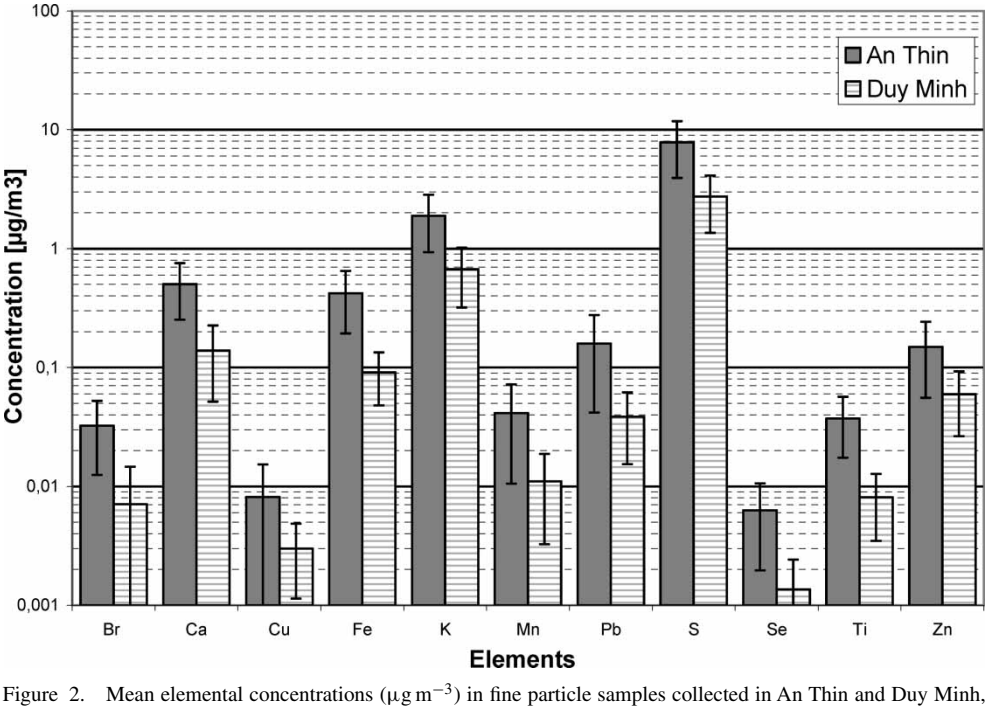

Figure 2. Mean elemental concentrations ($\mu g\,m^{-3}$) in fine particle samples collected in An Thin and Duy Minh, northern Vietnam, during November and December 2000. The error bars in the graph represent the standard deviation of elemental concentrations (number of samples: 15).

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areas [18]. Whereas the An Thin concentrations during this campaign were of the same order of magnitude as the mean values for the period April to October 2000, the concentrations measured in Ha Nam province were approximately two to three times lower than at the measurement sites in the Bac Ninh and Hai Duong provinces situated at a distance of 50–70 km east of Hanoi [11]. The higher concentrations of all analysed trace elements in the particle samples from An Thin are assumed to originate from the Pha Lai power plant, since the monsoonal wind patterns gave mainly north-easterly wind during the measurement campaign. This plant constitutes the main power supply for the northern part of the country with eight $220\,t\,h^{-1}$ coal-fired boilers and four 110-MW turbines [19]. The remarkable regional concentration differences between An Thin and Duy Minh support the finding that long-distance transport in South East Asia is much less efficient than at the mid-latitudes, because of low wind speeds and high amounts of rainfall in this part of the world [4].

The high Br concentrations in Vietnamese $PM_{2.5}$ suggest coal combustion and leaded gasoline as possible sources. The addition of anti-knock additives to leaded gasoline results in a Br/Pb mass ratio of 0.38 in fresh vehicle exhaust emissions. Thus, this ratio is often used as tracer of anthropogenic aerosol [20, 21]. Similarly as for Br, the most probable sources of the fine particulate Pb in Vietnam were coal combustion and automotive exhaust emissions from vehicles using leaded gasoline. The measured Br/Pb ratio of 0.2 at both sampling sites indicates either that exhaust emissions do not constitute the only source of Pb in fine particles or that a part of the emitted $PbBr_2$ may be rapidly oxidized to $PbSO_4$ in the warm and humid Vietnamese air [22]. Generally, the Vietnamese Pb concentrations in the fine particulate fraction were far below the limit of $5\,\mu g\,m^{-3}$ set in the Vietnamese air-quality standard. They even remained under the international WHO guideline of $0.5\,\mu g\,m^{-3}$ for particulate Pb [23, 24]. Both standards are based on 24-h averages and address the total concentration of airborne Pb.

A comparison of the mean S concentrations measured in An Thin during this campaign with concentrations in April to October of the same year [11] did not show any significant seasonal variations in sulfate concentrations for $PM_{2.5}$ particles. This observation is in agreement with a 2-yr data series measured in Kuala Lumpur during 1999–2000, where only a small seasonal variation in sulfate concentrations on $PM_{2.5}$ particles was found [25].

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3.2 *Elemental concentrations in freshwater and aquatic biota*

In figure 3, the mean elemental concentrations of freshwater samples from An Thin and Duy Minh are presented, as well as precipitation samples from An Thin. The concentrations of the elements As, Cr, Cu, Fe, Mn, Ni, and Zn in the investigated freshwater samples were below the Vietnamese legal limits listed for these elements in TCVN 5942-1995, with the exception of the mean Fe concentration in Duy Minh [26]. In contrast to the aerosol samples, the unambiguous trend of significantly higher concentrations in An Thin is not found in the water samples, where the mean concentrations of Cr, Fe, K, and Zn in Duy Minh exceeded those measured in An Thin.

Oxidized sulfur compounds are known to belong to the substances causing aqueous acidification effects by atmospheric input [27, 28]. The concentrations of total S of 12 and $5.2\,\mu g/ml$ measured in the freshwater samples from lakes situated in the neighbourhood of An Thin and Duy Minh, respectively, would correspond to equivalent sulfate concentrations of 36 and $16\,\mu g\,ml^{-1}$, respectively, if all S were to exist as sulfate. Such sulfate concentrations do not exceed the range of naturally occurring concentration levels [29]. The pH values of 6.5 and 6.8 measured in the water sampled in An Thin and Duy Minh, respectively, are close to neutral and do not reflect any acidification effect due to atmospheric sulfate input. To summarize, the results of the freshwater samples indicate that parameters other than atmospheric deposition

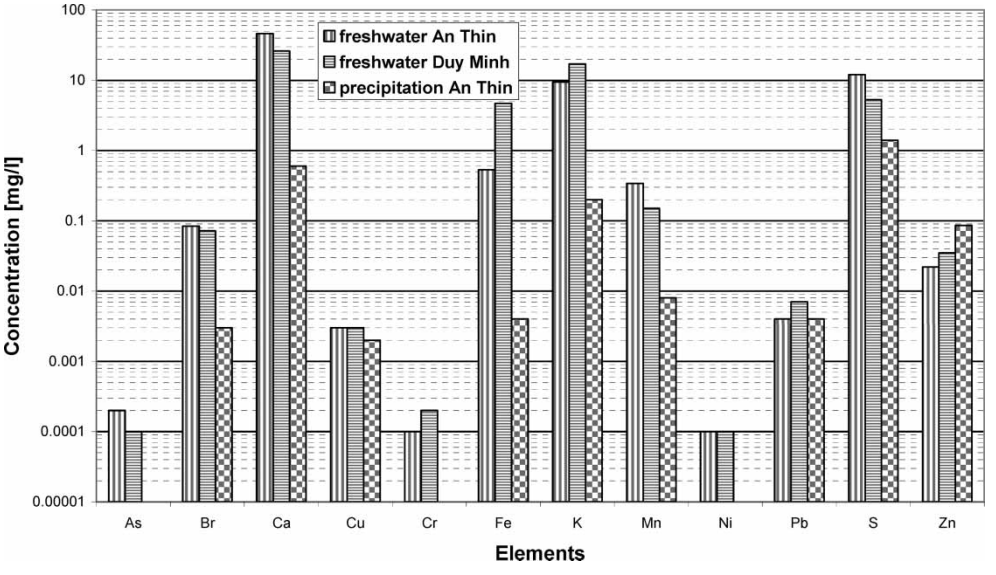

Figure 3. Mean elemental concentrations ($\mu g\,ml^{-1}$) in freshwater samples from An Thin and Duy Minh and rainwater samples from An Thin. Rainwater from Duy Minh was not available (number of samples: 2–3).

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may have a stronger impact on the elemental concentrations of the investigated freshwater samples.

In figure 4, the mean elemental concentrations and standard deviations of the analysed biomonitor tissues are presented. These tissues have been selected for this study, since biomonitors are in some aspects superior to water samples. They are able to bioaccumulate trace

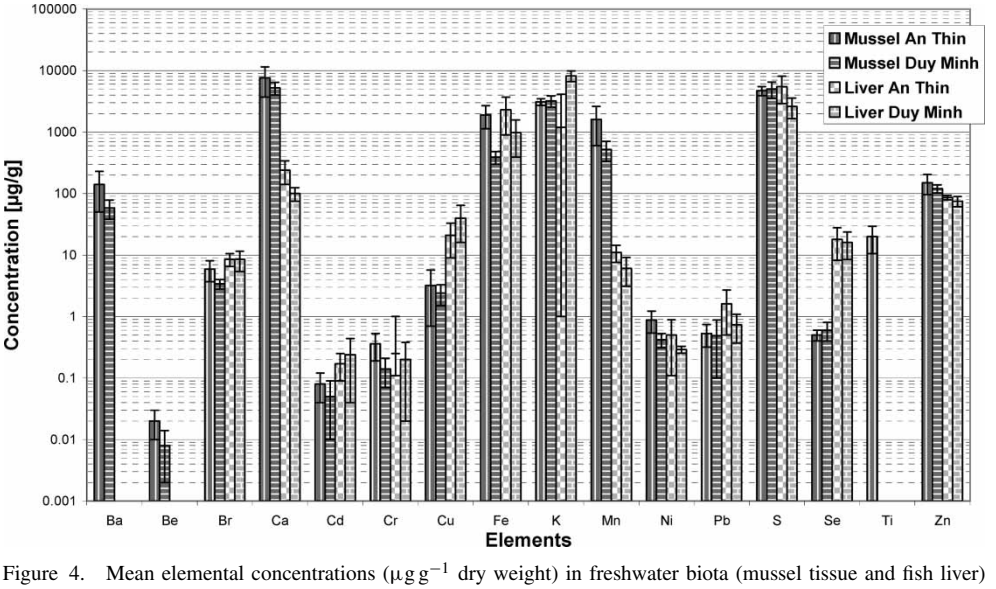

Figure 4. Mean elemental concentrations ($\mu g\,g^{-1}$ dry weight) in freshwater biota (mussel tissue and fish liver) from An Thin and Duy Minh. The error bars in the graph represent the standard deviation of elemental concentrations (number of samples: 10).

metals with concentration factors as high as 10^3–10^6 and provide a time-integrated measure of the ecotoxicologically important portion of a metal or substance in the respective aquatic environment [30].

Similarly, as for the freshwater samples, the unambiguous trend of the fine particle samples, i.e. significantly higher concentrations of all investigated elements at one site, was not reflected in the tissue types. A comparison between the two sampling sites (see figure 4) showed significantly higher concentrations in the An Thin mussel tissue for Ba, Be, Br, Cr, Fe, Mn, and Ni, whereas no element had significantly higher concentrations in Duy Minh. In the liver tissue, significantly higher concentrations were found in An Thin for Ca, Fe, Mn, Pb, S, and Zn, whereas the concentrations of Cr, Cu, and K were significantly higher in Duy Minh. The element Cr revealed an opposite trend in different tissue types, with significantly higher concentrations in the mussel tissue from An Thin and significantly higher concentrations in the liver tissue from Duy Minh. This emphasizes the important role of uptake pathways and cellular regulation mechanisms for the bioaccumulation of trace metals in biologic tissues.

A comparison of the elemental concentrations measured both in the Vietnamese mussel and fish samples with the International Standards compiled by the Food and Agricultural Organization (FAO) of the United Nations [31] showed that the concentrations of the elements addressed in these standards, i.e. Cu, Zn, As, Se, Cd, Pb, and Cr, were below the upper limits recommended for fish and fishery products in all investigated Vietnamese animal tissues. Compared with other biomonitoring studies using freshwater species, the concentration levels of most elements in the mussel and fish samples were of the same order of magnitude. The low levels of the trace metals Cd, Cu, Pb, and Zn at both sites indicated a comparatively unpolluted aquatic environment [12, 13].

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3.3 *Impact of atmospheric deposition on water and biota*

The comparative study of elemental concentrations in aerosol, freshwater, and biological samples collected in two different provinces in northern Vietnam revealed an evident discrepancy between air quality and aquatic biosphere. Whereas air quality was characterized by significant regional differences with considerably cleaner air in the agriculturally orientated Ha Nam province, the aquatic biosphere was relatively unpolluted at both sites, indicating a buffered impact of atmospheric deposition processes.

Elements of atmospheric origin can be deposited on terrestrial or aquatic surfaces by either dry or wet deposition. A number of climatic and meteorological parameters determine which of these processes is dominating [32]. For anthropogenic elements associated with fine particles, wet deposition is generally estimated to be more important than dry deposition, whereas crustal aerosols are characteristically associated with larger aerosols and are more efficiently removed by dry deposition [33]. The estimation of dry deposition fluxes of trace elements to water bodies is additionally complicated by micrometeorological parameters such as phoretic forces, electrostatic gradients, and water wave dynamics. Large uncertainties in flux estimates to water surfaces are caused by particle growth in the humidity gradient over water [34].

Considering these uncertainties, the deposition velocities (V_d) of trace elements in fine particles have been estimated using the mass median diameters (MMDs) suggested by Pirrone *et al.* [34] and the empiric equation based on particle-size distribution:

$$V_d = 0.388 \times \mathrm{MMD}^{0.76}$$

Table 2. Estimated deposition fluxes of trace elements in fine particulate matter ($PM_{2.5}$) in An Thin and Duy Minh during the air measurement campaign in November and December 2000.

Element	V_d (cm s^{-1})	Deposition flux An Thin (ng m^{-2} d^{-1})	Deposition flux Duy Minh (ng m^{-2} d^{-1})	Input difference (%)
Br	0.5	6.4	1.7	73
Ca	0.47	200	54	73
Cu	0.75	2.2	0.78	65
Fe	1.3	170	37	78
K	0.47	780	260	66
Mn	0.75	11	2.8	75
Ni	0.75	0.15	–	–
Pb	0.5	40	9.4	77
Se	0.5	1.6	0.33	79
S	0.23	1 600	520	66
Ti	1.3*	15	3.3	78
Zn	0.75	40	16	61

Note: The deposition velocity (V_d) was calculated using the mass median diameter for elements in fine particles suggested by Pirrone *et al.* [34] and the equation suggested by Davidson *et al.* [35]. The deposition fluxes were calculated by multiplying the mean ambient air concentrations presented in figure 2 by the estimated deposition velocities (V_d). The last column shows the input difference as a percentage between the two sites.
*The deposition velocity was set equal to that of the other crustal element.

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suggested by Davidson *et al.* [35] (table 2). The dry deposition fluxes (F) have been calculated by multiplying ambient air concentrations (C) by the estimated deposition velocities (V_d) (table 2):

$$F = V_d \times C.$$

The table shows that the estimated fine particle flux was 61–79% higher in An Thin for all investigated elements. This limited range (61–79%) of lower concentrations in Duy Minh suggests a major pollution source in the neighbourhood of An Thin. The estimates presented in table 2, however, are only based on the elemental concentrations in fine particles and thus underestimate the total dry deposition flux. It has been observed that dry deposition fluxes are determined by large particles, due to their high deposition velocity [28, 36]. Consequently, the elemental fluxes presented in table 2 are below the ranges of dry deposition fluxes presented by other workers [37]. They are meant to highlight regional differences rather than absolute fluxes.

As indicated by our results, the importance of atmospheric input is relative and strongly affected by the geographic characteristics of a water basin such as the ratio of water to land surface. Other factors influencing the absolute concentration increase caused by atmospheric input are sedimentation and evaporation. The amount of evaporation is influenced by the depth of a water body, with higher concentration increases in shallow lakes than in deeper lakes, as well as by climatic conditions [38]. The quantitative influence of these parameters is not known for the Vietnamese sampling sites.

Apart from the surroundings of the investigated water bodies, interactions with other deposited species might have weakened or counteracted the effect of some deposited pollutants. The sulfur concentrations in precipitation measured in An Thin during our campaign amounted to 1.4 mg l^{-1} in rainwater, see figure 3, as compared with the annual mean concentration of 0.96 mg/l sulfur observed in precipitation in Hanoi in 2000 [4]. Oxidized sulfur compounds belong to the substances causing aqueous acidification by atmospheric input [27]. The neutral pH values (pH $\sim$ 7) of the precipitation samples indicate that a possible acidification effect of sulfur-containing compounds might have been neutralized by atmospheric gases such as NH_3 or by the simultaneous deposition of alkaline compounds.

4. Conclusion

The results of this comparative study reveal that the mean concentrations of all investigated elements in fine particulate matter were significantly higher in An Thin, suggesting a strong local emission source in this area. The remarkable regional differences in air quality confirm the finding that long-range transport in South East Asia is not as efficient as in Europe, due to low wind speeds and a high amount of precipitation in this part of the world. In contrast to the particle samples, elemental concentrations in water and tissue samples were low at both sites and reflected a comparatively unpolluted aquatic biosphere. Significant site-specific concentration differences in the biologic samples were not as pronounced as for the particle samples and confined to specific elements and tissues. However, the significantly higher mean concentrations of a number of trace metals in An Thin mussels and liver tissue together with the deposition estimates suggest that the effects of atmospheric deposition are to some extent reflected in the elemental tissue concentrations. Our results indicate that the potential impact of elemental input by atmospheric deposition might have been affected by concomitantly deposited atmospheric bases as well as by biogeochemical characteristics of the investigated freshwater bodies. Long-term studies in the study area are required to investigate seasonal fluctuations in air quality and the environmental impact of deposition processes.

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Acknowledgements

This work has been financially supported by SIDA/SAREC, Sweden. We would like to thank Doan Viet Binh at the National Center for Natural Science and Technology in Hanoi for his careful collection of the samples and for useful biological advice, and Ngo Tra Mai for her assistance with the collection of air samples.

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