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NITROGEN COMPOUNDS IN WELL WATER AS A FACTOR OF A HEALTH RISK TO THE MACIEJOWICE COMMUNE INHABITANTS (MAZOWIECKIE VOIVODESHIP)

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Abstract: Well water quality monitoring was carried out in the Maciejowice Commune in the years 2005-2006. Water was sampled from 20 dug and drilled wells five times. Chemical analyses involved determination of NO_3^-, NO_2^-, NH_4^+, PO_4^{3-}, Cl^-, Ca^{2+}, Mg^{2+}, total hardness, pH and electrolytic conductivity. Health risk resulting from the presence of nitrates in water consumed by people was assessed in this paper. The obtained results indicated that 50% of examined waters did not meet the set standards. Ammonium ions and nitrates were the main ions contaminating drinking water. A negative correlation between nitrate, nitrite and ammonium ion concentration and well depth was found. People drinking water from 60% examined wells ingest excess quantities of nitrates (safety margin ADI: EDI < 1), which, in the case of long–term exposure, can be harmful, especially for infants and pregnant women. From among 20 analysed wells the water from 5 wells conformed to recommended standards. Its good quality was a result of the appropriate location of the wells within the household premises, appropriate depth and insulation and farmers' complying with the Code of Good Agricultural Practice.

INTRODUCTION

Groundwater is the source of drinking water for many people around the world, especially in rural areas [14]. Groundwater pollution is one of the most common environmental problems nowadays [5]. Numerous contaminants can render groundwater unsuitable for human consumption. Nitrate (NO_3^-) is the most frequently introduced pollutant into groundwater systems [20, 22]. Natural levels of nitrate in groundwater depend on soil type and geology. Crop fertilizers, domestic sewage and animal waste are common sources of nitrate contamination in rural areas [20, 29]. Nitrogen compounds in these sources are oxidized in aerated soil to soluble nitrate. Nitrate is highly water-soluble and therefore tends to migrate from soil to groundwater [18].

In Poland, rural domestic wells are especially vulnerable to nitrate contamination [11, 12, 15, 16, 23, 25]. Depending on the country regions, from 30 to 85% of these well intakes contain water with an exceeded concentration of nitrates [2]. High level of nitrate in drinking water is most often associated with shallow dug wells of less than 15 m in depth [11, 16].

Nitrate can have several adverse effects upon human health, most notably methemoglobinemia, gastric cancer and non-Hoadgkin's lymphoma [1, 4, 6, 7, 28]. The toxicology of nitrate (NO_3^-) to humans is mainly attributable to its reduction in the digestive system to harmful nitrite (NO_2^-) [1, 27]. Methemoglobin (MetHb) is formed when nitrite oxidizes the ferrous ion in hemoglobin (Hb) to the ferric form [4]. MetHb cannot bind oxygen, and the condition of methemoglobinemia is characterized by cyanosis, stupor, and celebral anoxia [4]. Under normal conditions, < 2% of the total Hb circulates as MetHb [4]. Raised levels of methemoglobin (greater than 10%) in infants under 4-6 months can produce cyanosis, referred to as "blue-baby syndrome" [1, 29]. In addition, nitrite can undergo nitrosation reactions in the stomach with amines and amides to form N-nitroso compounds (NOC) [27]. Most of these compounds are potent animal carcinogens, inducing tumors at multiple organ sites [26]. Therefore, nitrate concentration is an important criterion of drinking water quality [3, 17, 29]. To protect consumers from the adverse effects associated with the high nitrate intake, the World Health Organization (WHO), the European Union (UE) and Poland have set standards to regulate the nitrate concentration in drinking water. The drinking water standard set by the WHO [29], the UE [3] and Poland [17] for nitrate is 50 $mg \cdot dm^{-3}$.

The objective of the study was to determine nitrogen compound concentrations in well water and to assess health hazards which are associated with the nitrate presence in the drinking water of household wells in the Maciejowice Commune, Mazowieckie Voivodeship.

MATERIAL AND METHODS

Study area and sampling collection

Well water quality monitoring was carried out in the Maciejowice Commune in the years 2005-2006. The commune is located on the left bank of the River Wisła in the southern part of the Mazowieckie Voivodeship, and is a part of the Garwoliński administrative district. It is a predominantly agricultural commune where agricultural land makes up 50% and forested area 37% of the total area of the commune.

The water supply system in the Maciejowice Commune is currently under construction. By 2006, mains water was supplied to as little as 3% of the commune's households. As a result, the inhabitants predominantly utilize the water from shallow household wells. Studies included twenty wells being a source of water for both consumption and domestic purposes. Of the wells, sixteen were located in Maciejowice and four in Kochów; twelve wells were 3 to 50 m-deep drilled wells, and eight wells were dug wells whose depth ranged between 3.6 and 6 m; eleven wells were located in the high-density area (wells no 1-7, 9, 13, 14, 16), whereas the remaining wells were situated in the low-density area and surrounded by either cultivated fields (well no 10, 11, 17-20) or fallow land (well no 8, 12, 15). Water samples for testing were taken five times: in October 2005 and in April, May, June and October 2006.

Analytical procedures

Water samples for physical and chemical analysis were collected in polyethylene bottles and taken to the laboratory. Analysis was carried out immediately. The pH and electrical conductivity EC_{20} values were measured immediately after collection with the help of a portable pH-meter and conductivity - meter. Various colorimetric methods were used for the determination of NO_3^-, NO_2^-, NH_4^+, PO_4^{3-} [8, 13]. The concentrations of nitrates and nitrites were determined by the modified Griess method [13]. This method is based on the reaction of diazotization of the primary aromatic amines followed by coupling of diazonium salts yielding azo dye. Nitrite concentration was determined with sulphonamide and N-(1-napthyl) ethylenediamine. The same method was used for determination of nitrates after their reduction to nitrites. Amonium ion (NH_4^+) was determined by the indophenol blue method and PO_4^{3-} was determined by the molybdenum blue method. Total hardness, Ca^{2+} and Mg^{2+} were determined by the complexometric titration with EDTA and chloride (Cl^-) was determined by the argentometric method with silver nitrate [8].

Evaluation of a health risk

Health hazards associated with nitrate-contaminated drinking water were assessed by comparing Acceptable Daily Intake (ADI) with Estimated Daily Intake (EDI). The ADI value set for nitrates by the Joint FAO/WHO Expert Committee on Food Additives [6] is 5 mg $NaNO_3^-$·kg b.w.$^{-1}$·day^{-1}, which, when converted into NO_3^-, is 3.65 mg·kg b.w.$^{-1}$·day^{-1}. Following Cadum, Szczerbiński *et al.* [24] report the ADI value for nitrates (V) consumed with drinking water to be 0.365 mg NO_3^-·kgm.c.$^{-1}$·day^{-1}, that is 10% of the total ADI value consumed. Estimated Daily Intake (EDI) was calculated according to the formula:

EDI = F · R,

where:

F – average daily water consumption – 2 dm^3·person^{-1}·day^{-1},

R – concentration of nitrates in drinking water – mg·dm^{-3}.

Risk posed by nitrates in water was evaluated by determining a safety margin between ADI and EDI expressed as a value indicating how many times the daily intake of nitrates would have to increase to reach the ADI value, that is the value which, when exceeded, cannot be accepted as safe. The safety margin = ADI : EDI.

The following ranges were accepted following Szczerbiński *et al.* [24] on the basis of the safety margin calculated for nitrates in well water:

< 1 – below safety margin,
1–10 – low safety margin,
10–20 – medium safety margin,
20–30 – high safety margin,
> 30 – very high safety margin.

In the present work calculations were performed for an assumed human body weight of 70 kg.

Statistical analysis

Kolmogorov-Smirnov test was first used to check the normality of the data.

Because the data were non-normally distributed ($p < 0.05$), Spearman's rank order correlation was utilized to study associations between variables. The differences between concentrations of nitrate, nitrite and ammonium ions coming from various times of water sample collected, were estimated by nonparametric Kruskal-Wallis test. A value of $p < 0.05$ was considered statistically significant. Nonparametric methods have been found to be more accurate for the analyses of environmental data, especially groundwater data [9]. Statistical analysis was conducted using Statistica 5.0 computer program.

RESULTS AND DISCUSSION

Nitrate concentration in the well water tested ranged from 0.25 to 142.00 mg·dm^{-3} (Tab. 1). Throughout the studies the highest was the nitrate concentration in the water of well no 17 situated in Kochów. It was a very shallow well, 4.5 m in depth, located within the premises of a household whose wastewater was often disposed to the nearby meadow, despite the household possessing a septic tank. Nitrate concentration, ranging from 62.03 to 86.01 mg·dm^{-3}, was detected in the water of wells no 2, 3 and 14. They were dug wells situated in a high-density area and belonged to the households equipped with leaky septic tanks. In general, the standard value of 50 mg NO_3^-·dm^{-3} [17] was exceeded in the water of 25% examined wells. The lowest nitrate concentration, which did not exceed 10 mg NO_3^-·dm^{-3}, was found in the water of wells no 1, 9, 12, 16 and 18. The wells were drilled to the depth ranging from 7 to 50 m and situated within the household's premises at the appropriate distance from the house and livestock buildings. The households were equipped with properly situated and non-leaking septic tanks.

Table 1.Concentration of NO_3^-, estimated daily intake EDI and safety margin for nitrates taken with well water

Locality and well number	Concentration mg NO_3^- dm^{-3}		EDI mg NO_3^-·pers.$^{-1}$·day^{-1}	Safety margin ADI : EDI
	range	median	median	median
Maciejowice				
1	0.41-5.22	4.71	9.42	2.72
2	50.01-62.03	58.02	116.04	0.22
3	25.01-79.55	57.03	114.06	0.22
4	14.61-46.13	40.02	80.04	0.32
5	0.52-14.62	0.71	1.42	18.25
6	28.02-45.11	33.01	66.02	0.39
7	8.33-19.72	17.52	35.04	0.71
8	13.62-28.12	17.33	34.66	0.73
9	0.52-12.01	4.81	9.62	2.66
10	25.0-48.11	44.13	88.26	0.29
11	15.6-29.23	18.61	37.22	0.69
12	0.42-5.814	0.81	1.62	15.86
13	0.43-10.63	1.82	3.64	7.02
14	41.01-86.01	52.02	104.04	0.24
15	11.41-21.04	14.73	29.46	0.88
16	0.25-0.33	0.30	0.60	42.94
Kochów				
17	39.02-142.00	107.00	214.00	0.12
18	0.41-1.91	0.92	1.84	14.04
19	2.71-16.62	15.31	30.62	0.84
20	0.50-30.34	1.62	3.24	7.93

ADI- Acceptable Daily Intake; EDI- Estimated Daily Intake

Statistical analysis revealed a negative correlation between nitrate concentration and well depth (Tab. 2). It indicates that shallow dug wells are most likely to be nitrate contaminated. Significant relationships between well depth and nitrate concentration were confirmed by other authors too, e.g. Hudak [10], Jaszczyński *et.al.*[11], Nas and Berktay [14], Ostrowska and Płodzik [15].

Table 2. Spearman rank order coefficients r_s for the relationships between concentrations of NO_3^-, NO_2^-, NH_4^+ and some physical and chemical indexes of well water (n = 100)

Index	NO_2^-	NH_4^+	PO_4^{3-}	$CaCO_3$	Ca^{2+}	Cl^-	EC_{20}	Depth of well
NO_3^-	0.598**	ns	0.239*	ns	ns	0.519***	0.375***	-0.458***
NO_2^-		ns	0.207*	ns	ns	0.443***	0.374***	-0.356***
NH_4^+	ns		ns	0.326**	0.283**	0.263**	0.402***	-0.379***

*significant at $p < 0.05$; **significant at $p < 0.01$; *** significant at $p < 0.001$; ns- not significant

Health risk assessment showed that consumption by people of water from 60% examined wells was associated with nitrate intake which exceeded ADI, and as a result was below the safety margin (ADI : EDI < 1) (Tab. 1). The water of well no 17, whose ADI : EDI ratio was 0.12, was characterized by nitrate concentration which exceeded ADI eightfold. Long-term exposure to drinking water with such parameters increases the risk of digestive tract cancer [7, 28]. Nitrate intakes with drinking water representing the low safety margin (1–10), medium safety margin (10–20) and high safety margin (> 30) were associated with 20, 15 and 5% examined wells, respectively.

Nitrite concentrations in the water of wells sampled ranged from 0.00 to 9.80 $mg{\cdot}dm^{-3}$ (Tab. 3). The water of well no 14 exceeded the standard value of 0.5 mg $NO_2^-{\cdot}dm^{-3}$ [17] by as much as 20 times (9.80 mg $NO_2^-{\cdot}dm^{-3}$). In the close proximity of the well there was situated an organically-manured vegetable garden and a farmyard manure heap. Nitrogen is present in human and animal wastewater in organic form, which may then subsequently be mineralized to inorganic forms. The bacteria action on such organic matter results in its degradation and release of ammonia. The ammonia so produced may be oxidized to nitrite by bacteria such as *Nitrosomonas*, which can further be oxidized to nitrate by other bacteria such as *Nitrobacter*. These biologically mediated reactions are collectively reffered to as nitrification [21]. The nitrite ion contains nitrogen in a relatively unstable oxidation state. Chemical and bilogical processes can futher reduce nitrite to various compounds or oxidize it to nitrate. High nitrite concentrations may indicate that, among others, an intensive process of protein degradation is taking place. High dynamics of nitrate and nitrite concentrations in the examined well water are reflected in a statistically significant correlation between the ions concentrations (Tab. 2). There was also found a significant and negative correlation between nitrite concentration and well depth.

Ammonium ion concentration in the investigated well waters ranged from < 0.10 to 20.00 $mg{\cdot}dm^{-3}$ (Tab. 3). The highest ammonium ion concentration, within the range of 8.50 to 20.00 $mg{\cdot}dm^{-3}$, was found in the water of wells no 13 and 14. The standard value was exceeded by 0.5 mg $NH_4^+{\cdot}dm^{-3}$ [17] and was detected in the water sampled from ten wells. Such a high ammonium ion concentration indicates that there existed a nearby source of organic contamination, that is livestock buildings, farmyard manure heaps, and leaking septic tanks. Similar ammonium sources were mentioned in the studies by Ku-

Table 3. Range and median values of some physical and chemical indexes of well water

Index	Range	Median
NO_3^- (mg ·dm^{-3})	0.25–142.00	15.53
NO_2^- (mg ·dm^{-3})	0.00–9.82	0.05
NH_4^+ (mg ·dm^{-3})	< 0.10–20.00	0.45
PO_4^{3-} (mg ·dm^{-3})	0.10–20.00	1.11
Cl^- (mg ·dm^{-3})	5–110	35.5
$CaCO_3$ (mg ·dm^{-3})	88–654	255
Ca^{2+} (mg ·dm^{-3})	76.9–195.6	74.1
Mg^{2+} (mg ·dm^{-3})	3.6–46.8	14.6
pH	5.8–7.3	6.6
EC_{20} (μS·cm^{-1})	151–1500	549

czewski [12] and Szperliński [25]. A high ammonium ion concentration disqualifies the examined wells as sources of drinking water. There was found a negative correlation between ammonium ion concentration and well depth, which means that the shallower a well is the more leaking from natural manures and domestic waste occurs.

Low concentrations of ammonium ions and nitrites accompanied by high nitrate concentrations were determined for well no 17, which indicates that the contaminants covered a long way and in the meatime were subjected to the oxidation process.

Date of water sampling did not significantly differentiate nitrate, nitrite and ammonium ion concentrations, which was established by means of the nonparametric Kruskal-Wallis test ($p > 0.05$).

The problem of poor quality of well waters does not pertain exclusively to Maciejowice commune. Due to economic reasons or to a lack of pipelines, people in rural areas of our country often exploit dug and drilled wells which contain shallow ground waters. Numerous studies [11, 12, 15, 16, 23, 25] indicate that water from farm wells does not conform to the existing standards mainly because of high concentrations of nitrogen compounds. Concentrations of nitrites, ammonium ions and particularly of nitrates may exceed allowable standards several times.

Phosphate concentrations in the examined water samples were within a broad range of 0.10 to 20.00 mg·dm^{-3} with a median value of 1.11 mg·dm^{-3} (Tab.3). Phosphorus infiltration into groundwater may be associated with soil fertilization and livestock production but it may be as well introduced into the soil with detergents [19]. The presence of marked phosphorus concentrations in water may be indicative of the declining ability of the soil to bind this element.

Studies by Hudak [10] demonstrated that nitrates and chlorides are the most frequently infiltrating water contaminants. Chloride concentration in the water tested ranged from 5 to 110 mg·dm^{-3} with the median vale of 35.5 mg·dm^{-3} (Tab.3). There were no cases of the concentration exceeding the standard value of 250 mg·dm^{-3} [17]. Chlorides in well water come from livestock wastewater or domestic wastewater. Higher nitrate concentrations in the water of examined wells was accompanied by increased chloride concentrations. The relationship was confirmed by a statistically significant correlation between these indicators. Similar associations for well water were also reported by Jaszczyński *et* *al.* [11]. Zahn and Grimm [30] reported that chloride concentration in water is an indicator of the presence of nitrates.

The well water analysed was characterized by a varied degree of hardness; moderately hard and hard water constituted 35% in total. Water hardness depends on two cations, that is calcium and magnesium. There are no Polish standards for calcium concentration, whereas magnesium concentration should lie within the range of 30–125 $mg \cdot dm^{-3}$ [17]. The element was deficient in the water sampled from 90% wells studied. It is probably connected with the geological make-up of the terrain and low magnesium content of the rocks. Calcium concentration in the water samples ranged from 76.9 to 195.6 $mg \cdot dm^{-3}$ with the median value of 74.1 $mg \cdot dm^{-3}$ (Tab. 3).

The electrolytic conductivity in the water samples tested fell within the range of 151 to 1500 $\mu S \cdot cm^{-1}$ (Tab. 3) and at no occasion did it exceed the standard value of 2500 $\mu S \cdot cm^{-1}$ [17]. There was found a statistically significant correlation between the concentration of nitrogen compounds analysed and specific electrolytic conductivity, which indicates that the compounds influence the level of water mineralization.

It should be stressed that the overall sanitation status of well water tested was not satisfactory, which indicates that further studies are clearly warranted on a larger scale. The studies should be accompanied by an educational program to make people using contaminated water aware of potential risks associated with the poor quality of the water.

CONCLUSIONS

1. Nitrogen compound concentrations in 50% of examined well waters did not meet the set standards. Ammonium ions and nitrates were the main ions contaminating drinking water.
2. People drinking water from 60% examined wells ingest excess quantities of nitrates (safety margin < 1), which, in the case of long-term exposure, can be harmful, in particular for infants and pregnant women.
3. Water sampled from five wells met the drinking water standards throughout the whole study period. Its good quality was a result of the appropriate location of the wells within the household premises, appropriate depth and insulation and farmers' complying with the Code of Good Agricultural Practice.
4. Due to occurrence of non-standard concentrations of indicators affecting human health in well water as well as their marked dynamics, monitoring of well water quality ought to be carried out several times a year.

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ZWIĄZKI AZOTU W WODZIE STUDZIENNEJ JAKO CZYNNIK RYZYKA ZDROWOTNEGO MIESZKAŃCÓW GMINY MACIEJOWICE (WOJEWÓDZTWO MAZOWIECKIE)

W roku 2005-2006 na terenie gminy Maciejowice prowadzono monitoring wody studziennej. Badaniami objęto wodę pochodzącą z 20 studni kopanych i wierconych. Próby wody pobierano pięciokrotnie i oznaczono w nich stężenie: NO_3^-, NO_2^-, NH_4^+, PO_4^{3-}, Cl^-, Ca^{2+}, Mg^{2+}, twardość ogólną oraz pH i przewodność elektrolityczną. W pracy oszacowano ryzyko zdrowotne wynikające z obecności azotanów (V) w wodzie spożywanej przez ludność. Otrzymane wyniki wskazują, że stężenie związków azotu w 50% badanych wód studziennych nie odpowiada obowiązującym normom. Głównymi jonami zanieczyszczającymi wodę są jony amonowe i azotany (V). Wykazano ujemną korelację pomiędzy stężeniem azotanów (V), azotanów (III), jonów amonowych a głębokością studni. Oceniając ryzyko zdrowotne, stwierdzono, że ludność pijąca wodę z 60% badanych studni, pobiera azotany (V) w nadmiernej ilości (margines bezpieczeństwa ADI:EDI < 1), co w razie długotrwałości tego stanu może przynieść negatywne skutki zdrowotne, szczególnie dla niemowląt i kobiet w ciąży. Przez cały okres badań woda pochodząca z 5 studni spełniała zalecane normy. Dobra jakość tej wody wynikała z właściwej lokalizacji studni, a także przestrzegania przez rolników Kodeksu Dobrej Praktyki Rolniczej.