Hrvatski meteorološki časopis, 31, 89-94, 1996.

UDK: 551.588
Izvorni znanstveni rad

ON THE EMISSION AND REGIONAL BUDGET OF GREENHOUSE GASES IN HUNGARY

O emisiji i regionalnom budžetu stakleničkih plinova u Mađarskoj

A. MOLNÁR I E. MÉSZÁROS

Veszprém University, H-8201 Veszprém P.O.Box 158, Hungary

Primljeno 8. siječnja 1996, u konačnom obliku 15. lipnja 1996.

Abstract — Sources and sinks of atmospheric carbon dioxide, methane and nitrous oxide are estimated for the tropospheric box over Hungary. It is found that the total (biogenic and anthropogenic) strength of CO_2-C emission is around 100 $TgCyr^{-1}$.
16-20 % of this quantity is due to fossil fuel burning. This results in an annual anthropogenic emission of slightly less than 2 t per capita. The vegetation removes annually from the air over the country about half of the mass emitted, while a small part of carbon dioxide molecules goes to the stratosphere by diffusion. This means that our region is a net carbon source, even if combustion effects are neglected. The total methane release of the country lies around 500 $GgCyr^{-1}$ with the predominance of anthropogenic sources like solid waste treatment, natural gas production and distribution as well as animal husbandry. Taking into account appropriate numerical values, a chemical removal of 233 $GgCyr^{-1}$ can be calculated for the tropospheric air above the country. The sum of the magnitudes of the methane diffusion into the stratosphere and dry deposition is less nearly by an order of magnitude than the mass removed chemically. The comparison of sources and sinks indicates that the tropospheric box over the country is also a net source of carbon in reduced form. The emission of nitrous oxide (14 $GgNyr^{-1}$ for 1992) is controlled by the use of fertilizers, while a smaller amount is liberated during combustion. The quantity emitted is practically balanced by the stratospheric loss.

Key words: greenhouse gases, regional budget, Hungary.

Sažetak — Izvori i ponori atmosferskog ugljičnoga dioksida, metana i dušičnoga suboksida procijenjeni su za područje Mađarske. Utvrđeno je da je ukupna emisija CO_2-C (biogenetska i antropogenetska) oko 100 $TgCgod^{-1}$. 16-20 % toga iznosa posljedica je sagorijevanja fosilnih goriva, što čini godišnju antropogenetsku emisiju nešto manjom od 2 t po stanovniku. Vegetacija uklanja iz zraka oko polovicu emitirane mase godišnje, dok malen broj molekula ugljičnoga dioksida odlazi u stratosferu difuzijom. To znači da čak i nakon što se zanemare učinci sagorijevanja, to područje predstavlja neto izvor ugljika.
Ukupno oslobađanje metana jest oko 500 $GgCgod^{-1}$ s naglaskom na antropogenetske izvore, uklanjanje krutog otpada, proizvodnju i distribuciju prirodnog plina te stočarstvo. Primjenom odgovarajućih numeričkih vrijednosti, za troposferski zrak iznad promatranog područja, može se proračunati kemijsko uklanjanje metana: ono je oko 233 $GgCgod^{-1}$. Pri tome treba istaknuti da je ukupno uklanjanje metana iz troposfere procesima difuzije u stratosferu i suhog taloženja na tlo, za red veličine manje od uklanjanja kemijskim procesima. Usporedba izvora i ponora upućuje na zaključak da je i troposfera iznad područja Mađarske neto izvor ugljika u njegovu reduciranom obliku.
Emisija dušičnoga suboksida (14 $GgNgod^{-1}$ za 1992) najvećim dijelom potječe od upotrebe gnojiva, dok se manjim dijelom oslobađa u atmosferu sagorijevanjem. Emitirana masa dušičnoga suboksida praktički je uravnotežena gubicima u stratosferi.

Ključne riječi: staklenički plinovi, regionalni budžet, Mađarska.

1. INTRODUCTION

Atmospheric greenhouse gases play an important part in the control of the Earth`s climate. Since the concentration of these compounds is steadily increasing in the air, the study of their atmospheric pathways and budget is of particular interest. Greenhouse gases like carbon dioxide, methane and nitrous oxide have a rather long residence time in the atmosphere (several years) and consequently a spatial concentration homogeneity. For this reason the investigation of their global cycle is the most reasonable way of estimating perturbations due to human activities (see Rodhe, 1992).

However, regional scale budgeting can also be useful if we want to quantify the contribution of a given area (country) to the global cycle. This makes it easier to determine the possibilities of environmental management in the area or country considered. The solution of the global problem will be the result of such regional studies and mitigation measures.

The aim of this paper is to discuss the emission and removal of carbon dioxide, methane and nitrous oxide in Hungary and in the tropospheric air over the country. Thus, this work is an extension of our former publication on the subject (Mészáros and Molnár, 1992). One should mention that advective terms in the budget of this small box are not calculated since their magnitude is not comparable to the values of emission and removal rates. However, the comparison between emission and removal gives an estimate of the difference in material input and output by advection.

All the calculations in this work have been done for the years 1988-1992, when important changes in the economy of the country occurred. It is to be noted that the emission values presented here are slightly different from those submitted officially by Hungary for CORINAIR. However, these differences, owing to different data base and emission factors, do not alter the main conclusion of this paper.

2. THE CARBON DIOXIDE BUDGET

Carbon dioxide emission is caused essentially by *four sources:* direct release by vegetation, respiration of people and animals, liberation from soils and emission during fossil fuel combustion. *Release by vegetation* can be estimated on the basis of net primary production by assuming that 50 % of the assimilated carbon remains in the plants, while the other half is emitted (Fung et al., 1987). Considering the spatial distribution of different plants and forests, the Hungarian emission according to this source, is estimated to be 28.5 $TgCyr^{-1}$. About two thirds of this emission is due to agriculture-related activities. This amount has been found practically constant between 1988 and 1992.

Human and animal respiration can be determined by measuring the quantity of CO_2 exhaled, which is proportional to the weight of the body. The intensity of this source declined from 1988 to 1992 because of the decrease in live-stock in Hungary. The value for 1988 is 2.4 $TgCyr^{-1}$, while in 1992 it is equal to 1.8 $TgCyr^{-1}$.

The intensity of this CO_2 source is practically equally distributed among the respiration of men, cows and pigs.

Soil is also an important source of biological carbon dioxide. This source consists of three parts: microbiological activity in soil, root respiration and decomposition of organic matter on the surface. Considering the area of our country with different types of vegetation cover as well as emission factors as given in literature (Fung et al., 1987; Hampicke, 1980) the calculation results in a figure between 40-50 $TgCyr^{-1}$. This range is due to the different emission factors found in the references cited. The value of this source intensity is controlled mainly by deciduous forests and it remained stable during the time period under consideration.

Energy production is an essential anthropogenic CO_2 source. As Figure 1 shows the quantity of energy produced in Hungary decreased between 1988 and 1992 from 990 PJ to 798 PJ. Using information on the energy structure of the country (Statistical Yearbook, 1993) as well as the appropriate emission factors (Rotty, 1987), the calculations result in the total CO_2 emission due to fossil fuel burning represented in Fig.1. One can see that the strength of this carbon input decreased by 26 % from 1988 to 1992. Consequently, both energy and CO_2 emission per capita became lower over these five years. In 1988 the yearly carbon release by combustion per capita was 2.0 Mg, while is was only 1.5 Mg in 1992. However, even this latter value is greater than the world average, which is about 1 $Mgyr^{-1}$ (IPCC, 1990). It should be noted that during the same period the carbon dioxide concentration increased at a Hungarian background site which obviously reflects global influences.

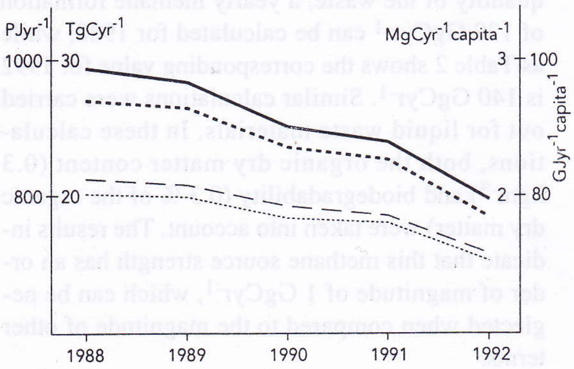

Figure 1. Energy production and carbon dioxide emission due to fossil fuel consumption in hungary from 1988 to 1992. per capita values are also plotted for these values.

Slika 1. Proizvodnja energije i emisija ugljičnoga dioksida uslijed sagorijevanja fosilnih goriva u Mađarskoj u razdoblju od 1988 do 1992. godine, i pripadni iznos emisije po stanovniku.

However, data indicate some decrease in the growth rate during the years considered in this study (Haszpra, 1994).

It should be noted that the cement industry and waste management also emit carbon dioxide into the atmosphere. Calculation indicates, however, that for Hungary, the contributions of these sources can be neglected compared to the release of CO_2 during energy production. Their intensity are below 0.5 $TgCyr^{-1}$.

As mentioned, *vegetation* is also a *carbon dioxide sink*. As with CO_2 released during respiration, the net sink can be determined on the basis of the mass of dry matter formed in plants (e.g. Bolin et al., 1977). Considering the appropriate data, a value of 55.0 $TgCyr^{-1}$ is calculated for the country. A negligible sink is provided by the removal of carbon dioxide by precipitation. However, this factor is less than 0.1 $TgCyr^{-1}$ for the tropospheric box over Hungary and thus it is not considered in the budget calculation. Considering the long tropospheric residence time of carbon dioxide, a certain part of its molecules penetrates the stratosphere from the box studied. This flux (F) can be determined by the following simple equation (Warneck, 1988):

$$F = K\rho \frac{m}{M} \frac{c}{h}$$

where K is the diffusion coefficient (= 1 m^2s^{-1}), r is the air density (0.37 kgm^{-3}), both at tropopause height at mid latitudes, m and M are the atomic mass of carbon dioxide and air, respectively, c is the mixing ratio at the tropopause level (3.5×10^{-4}), while h is an empirical scale height (at mid latitudes it is 25 km). By using this equation and the numerical values given and considering the area of Hungary (93,000 km^2), a stratospheric loss of 6.3 $TgCyr^{-1}$ is calculated.

Table 1 summarizes the above discussion. Data are tabulated for the year 1992.

First, we have to emphasize that the figures given should be considered with some caution. We estimate that the error in CO_2 emission by fossil fuel burning is about +/- 15 %; the uncertainty of the other terms reaches probably +/- 50 %. In spite of this, the values calculated suggest that atmospheric carbon dioxide generation vs removal is positive over Hungary: two times more oxidized carbon is released than removed. This means that even if energy production were entirely stopped, biogenic sources would produce more carbon dioxide than the amount removed from the air by vegetation. Unfortunately, it is not possible to determine the role of man in the unbalance of biological sources. Agricultural activities produce a large amount of carbon dioxide at present, but we do not know what the biogenic emission was before the begin-

Table 1. Hungarian carbon dioxide budget for 1992. The values are expressed in $TgCyr^{-1}$.

Tablica 1. Budžet ugljičnog dioksida za Mađarsku za 1992. godinu. Vrijednosti su izražene u $TgCgod^{-1}$.

Source/sink types	Source strength	Sink strength
Vegetation	28,6	57,2
People and animals	1,8	-
Soil	40,9-50,0	
Fossil fuels	15,1-16,6	
Stratospheric loss	-	6,3
Total	86,4-97,0	63,5

ning of intensive agriculture. Thus, this important question remains unanswered.

3. THE REGIONAL METHANE CYCLE

To estimate the Hungarian methane emissions we first considered animal husbandry. The methane production can be calculated taking into account the number of ruminants and the quantity of foodstuff consumed by them. On this basis we have found that in the country the methane production from fermentation of ruminants was 130 $GgCyr^{-1}$ in 1988. The corresponding figure in 1992 was 93 $GgCyr^{-1}$. This variation is due to the reduction in cattle number caused by important changes in animal husbandry.

Soil and water surfaces also emit CH_4 molecules, mostly in marshy area. This value has been determined by using information on the nature of the surface of the country and on the emission factors available (Ehhalt, 1985; Holzapfel-Pschorn and Seiler, 1986). Calculations indicate that the intensity of this source is somewhat less than that resulting from animal husbandry and its value decreased from 53-92 $GgCyr^{-1}$ to 50-84 $GgCyr^{-1}$ between 1988 and 1992 (see Table 2). This decrease was caused by some reduction in rice production, which is not an important branch in Hungarian agriculture.

Methane is also released into the air from *solid* and *liquid waste materials*. According to Bingemer and Crutzen (1987) under anaerobic conditions about 80 % of the organic matter is transformed and results in biogas, 50 % of which is CH_4. Such circumstances are created if solid wastes are deposited in landfills or open dumps. Taking into account this information and the composition and quantity of the waste, a yearly methane formation of 130 $GgCyr^{-1}$ can be calculated for 1988, while as Table 2 shows the corresponding value for 1992 is 140 $GgCyr^{-1}$. Similar calculations were carried out for liquid waste materials. In these calculations, both the organic dry matter content (0.3 kgm^{-3}) and biodegradability (0.3 % of the organic dry matter) were taken into account. The results indicate that this methane source strength has an order of magnitude of 1 $GgCyr^{-1}$, which can be neglected when compared to the magnitude of other terms.

The main component of natural gas used for energy production is methane. Due to this fact, during the *mining and distribution of natural gas*, some methane is released into the air. According to literature (CONCAWE, 1986) and information given by the Hungarian Gas Industry Trust, methane release during mining is equal to 2 %. Considering the quantity of natural gas exploited in Hungary this results in a yearly methane emission of 58 GgC and 46 GgC for the years 1988 and 1992, respectively. Methane is also liberated at joining points of pipe-lines during natural gas distribution. In the CONCAWE report, the loss is 3 % of the amount of gas. Taking into account the amount of natural gas distributed in the country a value of 142 $GgCyr^{-1}$ can be calculated for 1992. This is by 12 % lower than the emission in 1988 (see Table 2).

Coal and lignite mining also provides a methane source, since coal contains about 5 m^3 of CH_4 per ton, 50-100 % of which comes into the air during mining. For the methane content of lignite, we estimated the same value and assumed that 25-75 % of this methane escapes into the air (Ehhalt, 1985).

Table 2. Hungarian methane emission (in $GgCyr^{-1}$) from 1988 to 1992. The ranges given demonstrate the uncertainty of the calculations.

Tablica 2. Emisija metana (u $GgCgod^{-1}$) za Mađarsku 1988.—1992. Raspon pokazuje nepouzdanost proračuna.

Sources	1988	1989	1990	1991	1992
Animals	130	123	119	110	93
Soil	53-92	53-90	53-90	51-85	50-84
Coal mining	28-60	26-56	20-44	19-42	17-38
Natural gas	220	226	198	207	188
Solid waste	130	130	130	120	140
Total	561-632	558-625	520-581	507-564	488-543

The results are also tabulated in Table 2. They indicate an important reduction during the time period under investigation owing to the decline of coal mining in Hungary.

The *major sink of methane* in the troposphere is its removal by chemical reaction with OH radicals. Preliminary results of methane monitoring in Hungary indicate (Haszpra, personal communication) that the average CH_4 concentration in the background surface air is around 1.9 ppm. This is a bit higher than the global tropospheric average (1.7 ppm). We assumed that in the tropospheric air box over the country the average methane level is between these two values and equal to 1.8 ppm. Taking into account this concentration, at a tropopause height of 12000 m and at a rate constant of 7.7×10^{-15} $cm^3 molecule^{-1} s^{-1}$ (Warneck, 1988), a chemical removal of 233 $GgCyr^{-1}$ can be calculated for the air above the country.

A part of methane molecules *escapes into the stratosphere* by diffusion. The flux of methane mixed into the stratosphere can be determined by the equation used above for carbon dioxide and a mixing ratio of 1.7×10^{-6}. In this way a stratospheric loss of 30 $GgCyr^{-1}$ can be determined.

To complete our calculation, the *dry deposition* of methane molecules was also considered. On the basis of the total dry deposition given in IPCC (1990) a dry deposition velocity of 1.6×10^{-6} ms^{-1} can be estimated. Taking this figure and a concentration of 1.9 ppm for the surface air over Hungary, one can calculate a total yearly dry deposition of a magnitude of 4.4 GgC that can be practically neglected considering other sink values.

Summing up the above sink amounts we can say, with caution, that the total methane mass removed from the tropospheric box over Hungary is yearly equal to 267 GgC. Comparing this figure with the source values in Table 2, one can conclude that the box over Hungary is a net methane source. In other words, the methane input into the box by advection is much smaller than the output. The difference between "export" and "import" is at least 300 $GgCyr^{-1}$. Further research is needed, however, to confirm this conclusion.

4. NITROUS OXIDE

Nitrous oxide is the most abundant nitrogen compound in the atmosphere. This greenhouse gas is formed in the soil by nitrification and denitrification. The most direct way of N_2O formation is by reduction of nitrate ions by hydrogen in gaseous and ion form. However, nitrifying micro-organisms also produce N_2O during the nitrification of ammonium and hydroxilamine, both coming from the decomposition of organic materials (Brenner and Blackmer, 1981). The emission from different soils treated with nitrogen-fertilizers in the form of ammonium sulfate or urea is particularly significant.

The release of N_2O from soil not treated with fertilizers can be estimated on the basis of the data supplied by Fenger et al. (1990) giving the nitrous oxide emission for unit area of different soils. Using this procedure a value of 1.2 Gg of annual nitrogen emission has been calculated for forest and grassland in Hungary for each of the years studied. Taking into account the relationship between N_2O emission and the quantity of different N-fertilizers used (Fenger et al., 1990), the calculation results in a value between 12.7 (1988) and 10.6 (1992) $GgNyr^{-1}$.

A certain amount of N_2O is also emitted from *combustion sources* during fossil fuel burning. By applying statistical information on the quality and quantity of different fossil fuels consumed in Hungary as well as the emission factors proposed by Fenger et al. (1990) values in the range of 1.3-1.7 $GgNyr^{-1}$ and 1.4-1.7 have been obtained for stationary and mobile sources, respectively. The lower end of the range refers to 1992, which indicates a slight reduction in this source over five years.

The N_2O-N emissions from different Hungarian sources are summarized in Table 3. One can see

Table 3. Nitrous oxide emission (in $GgNyr^{-1}$) in Hungary for 1992.

Tablica 3. Emisija dušičnoga suboksida (u $GgNgod^{-1}$) za Mađarsku za 1992. godinu.

Source type	Source intensity
Fossil fuel combustion	1,4
Mobile sources	1,3
N-fertilized soil	10,5
Non-fertilized soil	1,2
Total	14,4

that liberation from fertilized soil gives 74 % of the total emission, while combustion sources contribute 19 % of the total. This means that the major part of nitrous oxide emission in the country is due to human activities.

Our knowledge of the *sinks* of nitrous oxide on the surface is rather uncertain. For this reason it is usual to neglect the dry deposition of this gas (IPCC, 1990). On the other hand, one believes that N_2O is chemically stable in the troposphere and it goes into the stratosphere by diffusion. This stratospheric loss over Hungary was calculated by using the method outlined for CO_2 by assuming a nitrous oxide concentration of 0.31 ppm (Warneck, 1988). In this way we calculated a sink term of 13.0 $GgNyr^{-1}$, which is practically equivalent to the total source strength (see Table 3). Of course, these results should be considered with caution because of the uncertainties involved in the calculation. In spite of this, it seems that the stratospheric loss over the country balances the emission. In other words the difference in advection terms is equal to zero.

5. CONCLUSION

The emission calculations presented above show that between 1988 and 1992 the emission of greenhouse gases decreased in Hungary. This reduction was caused by changes in energy production and agriculture. In spite of this reduction the release of carbon dioxide and methane into the air is higher than their removal by sink processes. It seems that stratospheric loss over the country balances the nitrous oxide emission.

REFERENCES

Bingemer H.G. and P.J. Crutzen, 1987: The production of methane from solid wastes. *J. Geophys. Res.*, **92**, 2181-2187.

Bolin B., E.T.Degens, S. Kempe and P. Ketner, 1977: (eds.) *The global carbon cycle.* SCOPE 13. John Wiley ans Sons Ltd., Chichester-New York-Brisbane-Toronto, 337-345.

Brenner J.M. and A.M. Blackmer, 1981: Terrestrial nitrification as a source of atmospheric nitrous oxide. In *Denitrification, Nitrification and Atmospheric Nitrous Oxide* (ed. C.C. Delwicke), John Wiley ans Sons, Inc., New York, 151-169.

CONCAWE: *Volatile organic compound emissions: an inventory for Western Europe.* Report No 2/86. 1986. Den Haag.

Ehhalt D.H., 1985: Methane in the global atmoshere. *Environment*, **27**, 10.

Fenger J., J. Fenhann and N. Kilde, 1990: *Danish Budget for Greenhouse Gases.* Nordic Council of Ministers, Copenhagen.

Fung I.Y., C.J. Tucker and K.C. Prentice, 1987: Application of advanced very high resolution radiometer vegetation index to study atmosphere-biosphere exchange of CO_2. *J. Geophys. Res.*, **92**, 2999-3015.

Hampicke U., 1980: The effect of the atmosphere-biosphere exchange on the global carbon cycle. *Experiencia*, **36**, 776-780.

Haszpra L., 1994: Carbon dioxide concentration measurements at a rural site in Central Europe. *Tellus*, **46B**, (in press).

Holzapfel-Pschorn A. and W. Seiler, 1986: Methane emission during a cultivation period from an Italian rice paddy. *J. Geophys. Res.*, **91**, 11803-11814.

IPCC (Intergovernmental Panel on Climate Change) (1990) *Climate Change* (eds: J.T. Houghton, G.J. Jenkins and J.J. Ephramus). Cambridge University Press, Cambridge.

Mészáros E. and A. Molnár, 1992: Energy production, economy and greenhouse gas emissions in Hungary. In *Global Climate Change: Implications, Challenges and Mitigation Measures* (eds.: S.K. Majumdar, L.S. Kalkstein, B.M. Yarnal, E.W. Miller and L.M. Rosenfeld). Publ. Pennsylvania Academy of Science, Easton, PA., 471-480.

Rodhe H., 1992: Modeling biogeochemical cycles. In *Global Biogeochemical Cycles* (eds.: S.S. Butcher, R.J. Charlson, G.H. Orians and G.V. Wolfe), Academic Press, London, San Diego, New York, Boston, Sydney, Tokyo, Toronto, 55-72.

Rotty R.M., 1987: A look at 1983 CO_2 emissions from fossil fuels (with preliminary data for 1984). *Tellus*, **39B**, 203-208.

Statistical Yearbook 1992, 1993: Hungarian Central Statistical Office, Budapest.

Warneck P., 1988: Chemistry of the Natural Atmosphere. Academic Press, San Diego, New York, Berkeley, Boston, London, Sydney, Tokyo, Toronto.