GAIA

ÖKOLOGISCHE PERSPEKTIVEN FÜR WISSENSCHAFT UND GESELLSCHAFT **ECOLOGICAL PERSPECTIVES FOR SCIENCE AND SOCIETY**

4 | 2013



- HUMAN IMPACT ON EARTH SYSTEMS
- SHARING ECONOMY
- ANPASSUNG AN DEN KLIMAWANDEL



Reducing the Carbon and Sulfur Load of the Atmosphere

Assessing Global Decarbonization and Desulfurization and the Future of a Low-Carbon World

During the past century carbon and sulfur concentrations in the atmosphere have increased steadily. Great efforts have been made to lower human-related emissions: on a global scale we observe absolute desulfurization but only relative decarbonization. Further reductions are imperative, but it seems unlikely that the dependence on carbon will end soon.

Vaclav Smil

Reducing the Carbon and Sulfur Load of the Atmosphere. Assessing Global Decarbonization and Desulfurization and the Future of a Low-Carbon World *GAIA* 22/4 (2013): 255–262

Abstract

For more than a century human activities have been affecting two key biogeochemical cycles, carbon and sulfur. Substantial declines in average carbon intensity (carbon per gigajoule), especially in Western countries, have not been translated into an absolute decline of global emissions, largely due to large emissions increases in Latin America, Africa and, above all, in Asia. In contrast, global desulfurization has taken place not only in relative but also in absolute terms: we have seen not only lower sulfur intensities per unit of liberated energy but substantial decline in total sulfur emissions. We have accomplished this despite a massive increase in global combustion of coal thanks largely to an effective technical fix (flue gas desulfurization). There are no imminent prospects for any major reductions in absolute emissions of carbon dioxide. In view of climate change, this seems critical because the atmospheric behavior and the future levels of absorbed outgoing radiation are indeed determined by the absolute atmospheric levels of the gas, not by reductions of relative burdens per unit of product or service. The next two decades will prove to be crucial.

Keywords

decarbonization, desulfurization, emissions, fossil fuels, hydrogen economy, primary electricity, primary energy, sustainability

Contact: Dr. Vaclav Smil, Distinguished Professor Emeritus | University of Manitoba | Department of Environment and Geography | Winnipeg, MB R3T 2M6 | Canada | Tel.: +1 204 4749451 | E-Mail: vsmil@cc.umanitoba.ca

© 2013 V. Smil; licensee oekom verlag.

This is an article distributed under the terms of the Creative Commons Attribution

License (http://creativecommons.org/licenses/by/3.0), which permits unrestricted use,

distribution, and reproduction in any medium, provided the original work is properly cited.

ombustion of fossil fuels has been the most important cause of increasing atmospheric concentrations of carbon dioxide (CO₂) and of the rising emissions of acidifying compounds, two major components of global environmental change (Hester and Harrison 2002, Mackenzie 2011). Carbon is the dominant constituent of phytomass (wood, crop residues) and fossil fuels (coals and hydrocarbons). Its rapid oxidation is by far the largest source of thermal energy released during combustion. And while wood has a very low sulfur content, most coals, as well as many crude oils and some natural gases, contain enough sulfur (commonly one to two percent by mass) to produce relatively large amounts of sulfur dioxide (SO₂) (Smil 2008).

The first concerns about acidification of aquatic ecosystems date to the late 1960s (Royal Ministry of Foreign Affairs 1971). During the 1970s and the 1980s acid rain (more accurately acid deposition, wet and dry) was commonly perceived as one of the most challenging – as well as one of the most worrisome – environmental problems affecting both aquatic and terrestrial ecosystems and damaging human health and materials (Rhode 1989, Smil 1997) (figure 1, p. 256). Subsequently, anthropogenic global warming has emerged as the leading environmental challenge, receiving an enormous amount of scientific and public attention (IPCC 1990, 2007, Nordhaus 2008).

Concerns about human interference in global carbon and sulfur cycles have been translated into explicit efforts to lower the atmospheric emissions of CO_2 and SO_2 . The eventual goal is to supply all of the needed energy by a new system devoid of carbon and sulfur. But our quest to lower these emissions predates the concerns about global environmental change. I will show how a combination of fuel substitutions and technical advances – pursued for a variety of reasons, ranging from economic advantages to technical opportunities – has led to a significant *relative decarbonization* of modern energy systems on both national and global scales and, even more impressively, to *absolute desulfurization* of the global energy system.

There are four fundamental strategies to reduce emissions of CO₂ and SO₂ in any energy system: to use energy more efficient-

ly, i.e., to derive more useful services (heat, light, motion) from every unit of fuel and primary electricity; to burn fuels that contain less carbon and sulfur; to capture generated gases (CO₂ and SO₂) before they are released into the atmosphere; and to rely more on commercially available non-carbon energy conversions. Using energy more efficiently is the most desirable general path toward more rational energy futures. This option has received a great deal of attention during the past four decades. In this brief paper I will therefore focus on the other three specific options.

RESEARCH

The substitution of solid fuels by hydrocarbons – the first option – has been the key process combining gradual decarbonization and desulfurization of modern energy supply: refined liquid fuels and natural gases have higher shares of hydrogen than wood and coal and lower shares of sulfur than most coals. Their combustion results in lower specific emissions of carbon and sulfur per unit of liberated energy. In the past this transition has not been driven by any explicit quest for decarbonization but rather by a deliberate effort to introduce higher-quality fuels 1. that have

FIGURE 1: Combustion of fossil fuels has been one of the most important causes of the rising emissions of acidifying compounds. The accelerated decay of sculptures made of limestone and marble impressively demonstrates the detrimental effects of acid rain.



higher energy densities which can be converted to useful energy with higher efficiencies, 2. that are easier to handle, and 3. that generate less particulate matter (soot, fly ash) per unit of energy. In contrast, lower emissions of SO₂ from hydrocarbons were often an important, even the dominant, consideration leading to substitutions, first in order to improve air quality of cities whose inhabitants suffered from exposure to classical (particulates and SO₂) smog, later in order to reduce acid deposition.

Post-combustion removal of CO_2 – the second option – has emerged as a possible (albeit very expensive) technical option during the 1990s, but the progress toward commercial carbon capture and sequestration (CCS) has been slow (Metz et al. 2005, Global CCS Institute 2013) (figure 2). Again, in contrast, installation of the first flue gas desulfurization (FGD) units in large coal-fired electricity-generating plants began during the late 1960s. By 1999 almost 30 percent of the United States (US) coal-fired electricitygenerating capacity had some sulfur emission controls. Today FGD is a standard installation in all large power plants burning sulfur-containing fuel (Hudson and Rochelle 1982, USEPA 2013 a). Most importantly, since 2000 China has far surpassed the US in total FGD capacity: by 2009 McIlvaine's inventory identified FGD installed on 379 gigawatts of generating capacity compared to only 130 gigawatts in the US (McIlvaine 2009).

Increasing reliance on non-carbon (and hence also non-sulfur) alternatives - the last approach - is an ancient option but traditional water wheels and windmills made only limited contributions (Smil 2008). Only modern primary (non-carbon-based) electricity generation has made a notable difference: hydroelectricity has been generated on an increasing scale since the 1880s; geothermal plants made modest contributions since the early 20th century; nuclear fission entered during the late 1950s; wind turbines and solar photovoltaics began their ascent during the 1990s. Compared to these conversions, geothermal and solar energy used for water and space heating is a minor contribution. I will quantify the contributions of all of these shifts for the entire 20th century and for the first decade of the 21st century, and will do so at the global level as well as for four leading protagonists: the US, France, Japan and China.

Decarbonization

Fuel Substitutions: From Wood to Natural Gas

Wood, the dominant fuel in all pre-industrial societies, is composed mostly of three biopolymers: cellulose, hemicellulose, and lignin (Smil 2013). Cellulose – $(C_6H_{10}O_5)_n$ – is a linear polysaccharide and the biosphere's most ubiquitous strong structural macromolecule while hemicelluloses produce essentially an amorphous and structurally weak biopolymer. Lignin is a complex polymer of conyferyl, coumaryl and synapyl alcohols and its linkages with hemicelluloses stiffen all woody phytomass. Carbon makes up 44.4 percent of cellulose, about 45 percent in hemicelluloses, values for lignin range between 61 to 66 percent (Sjöström 1993, USDA 2010).



FIGURE 2: Coal-fired electricity-generating stations are the world's largest point sources of both CO₂ and SO₂. Jänschwalde, near Peitz in Brandenburg (very close to the Polish border), is Germany's largest lignite-fueled station with three gigawatts of installed electrical capacity; it generates annually 22 terawatt-hours of electricity and it was to be the site of a large-scale carbon capture and sequestration project that was cancelled in 2011.

Analyses of many wood species (all in terms of dry weight) show the median values for lignin, cellulose and other biopolymers at, respectively, 26, 46 and 21 percent for deciduous trees and 29, 44 and 23 percent for conifers (Cornwell et al. 2009). Carbon content of wood containing 25 percent lignin and 75 percent cellulose and hemicelluloses would be almost exactly 50 percent. This value is commonly used in approximate conversion of woody phytomass to carbon. Actual carbon shares (all values are for dry wood) range between 46 to 50 percent (average about 48 percent) for hardwoods, and (due to their higher lignin content) 47 to 55 percent (average about 51 percent) for conifers (Lamlom and Savidge 2003, USDA 2010) while hydrogen accounts only for about six percent. For coal, typical mass shares of the two elements are 65 percent carbon and five percent hydrogen, liquid fuels refined from crude oil have 86 percent carbon and 13 percent hydrogen. Methane (CH₄, the dominant constituent of natural gas) has 75 percent carbon and 25 percent hydrogen by mass.

The first impression might be that replacing wood by coal will not help to decarbonize energy supply: when using averages of 50 percent carbon and six percent hydrogen, wood would have an atomic hydrogen-carbon ratio (H:C ratio) of about 1.4 compared to 1.0 for typical bituminous coal. But that is not the ratio of

atoms whose combustion is converted to thermal energy: a large part of wood hydrogen is never oxidized because hydroxyl (OH⁻) radicals that are part of cellulose and hemicellulose escape to the air during the early stages of combustion (Shafizadeh 1981). That is why Ausubel and Marchetti (2007) argued that the true H:C ratio for wood should be just 0.1. In reality, the ratio is variable because temperate and tropical wood species also contain organic extractives, from less than ten up to 20 percent respectively (USDA 2010). Hydrogen in these fats, waxes, resins and essential oils is oxidized during combustion but effective H:C ratios in wood combustion are still just small fractions of coal's 1.0.

Shifting from bituminous coals and lignites to hydrocarbons raises hydrogen's contributions substantially. Automotive gasoline and aviation kerosene, the two leading liquid fuels refined from crude oil, have a H: C ratio of 1.8 and pure methane has a H: C ratio of 4.0; actual ratio for natural gases is marginally lower due to the presence of higher homologs, ethane (C_2H_6) and propane (C_3H_8), besides methane. In terms of typical specific carbon emissions the declining progression – with default rates taken from IPCC (2006) – is from 30 kilograms of carbon per gigajoule for wood (energy density between 16 to 19 gigajoules per tonne) to almost 28 kilograms of carbon per gigajoule for poor lignites

(energy density less than 15 gigajoules per tonne), about 26 kilograms of carbon per gigajoule for good bituminous coal (25 to 26 gigajoules per tonne), 20 kilograms of carbon per gigajoule for refined liquid fuels (42 to 44 gigajoules per tonne) and to 15.3 kilograms of carbon per gigajoule for methane (energy density 53.6 gigajoules per tonne or 38.7 megajoules per cubic meter).

RESEARCH

Reasonably good historical statistics for the US (Schurr and Netschert 1960) and France (Barjot 1991) allow us to follow the first wave of decarbonization, from wood to coal. Wood supplied 91 percent of all fuels in the US in 1850 and 21 percent by 1900. The average emission rate declined from 29.6 to 24.7 kilograms of carbon per gigajoule, the latter value reflecting still low but rising contribution of hydrocarbons. In France – where wood supplied 76 percent of all fuel energy in 1850 and 32 percent in 1900, with virtually no oil or gas - the rate fell from 28.9 to 26.4 kilograms of carbon per gigajoule. By 1950 fossil fuels had reduced wood to marginal shares in all affluent countries (by 1950 it supplied just 4.5 percent of all fuel energy in the US, 13 percent in France). As expected, due to rising hydrocarbon extraction the decarbonization of fuel supply progressed much faster in the US (average emission rate of 21 kilograms of carbon per gigajoule in 1950) than in Europe (1950 mean of 24.6 kilograms of carbon per gigajoule in France) where imports of crude oil took off only during the 1950s. This means that between 1850 and 1950 the carbon intensity of fuel combustion decreased by 15 percent in France but by nearly 30 percent in the US.

In Japan, where economic modernization began only after the Meiji restoration during the 1880s, wood supplied still 52 percent of all fuel energy in 1900 and about 14 percent in 1950 when coal was the dominant fuel (SB 2013); the average emission rate was lowered from 28.3 to 25.2 kilograms of carbon per gigajoule during those 50 years. In China traditional biofuels (wood and straw) provided about 85 percent of all fuel energy in 1950 (down from 94 percent in 1900) and still 28 percent in 1980 when the country embarked on its rapid modernization energized mostly by coal (Smil 1988). China's average fuel emission rate had hardly changed between 1900 and 1950 - it remained above 29 kilograms of carbon per gigajoule. It was still close to 26 kilograms of carbon per gigajoule in 1980.

In the year 1950 the US were the only major economy with a large share of hydrocarbons but post-World War II economic expansion brought increasing oil imports to Europe and Japan. Gas imports (pipelines and liquefied natural gas) began during the 1960s. As a result, carbon intensities of fuel combustion decreased in all affluent economies previously dependent on coal: between 1950 and 2010 these declines were 15 percent in the US, 26 percent in Japan, and 27 percent in France. But rising oil and gas consumption in post-1980 modernizing China was swamped by huge increases in coal extraction – it had more than quintupled in 30 years and the carbon intensity of the country's fuel combustion declined by only five percent (to 24.2 kilograms of carbon per gigajoule) between 1980 and 2010 (LBNL 2013). Not surprisingly, global average shows much slower shifts because traditional biofuels retain their importance in many low-income countries and because China and India remain highly dependent on coal. As a result, global carbon intensity of fuel supply declined by only about 15 percent during the 20th century and by just additional six percent between 2000 and 2010.

Non-Carbon Energies: Primary Electricity

Carbon intensity of the total primary energy supply (TPES) falls as an increasing share of total energy consumption comes from primary electricity generated either by conversions of renewable flows or by nuclear fission. Hydroelectric generation became the first non-carbon source of energy beginning in the 1880s; in many countries it remained the only - or overwhelmingly dominant renewable conversion for the next 100 years. Everywhere its contributions expanded notably only after World War II (Smil 2008). Commercial exploitation of nuclear fission started during the late 1950s and it experienced its fastest growth during the 1970s (WNA 2013). In 1970 global hydro generation was more than 15 times larger than nuclear generation, by 2010 it was less than 25 percent higher (BP 2012). Geothermal electricity has remained a negligible contributor for decades, but in some countries wind turbines and photovoltaic cells have become important sources of non-carbon electricity. Even so, their contribution remains modest: in Europe, where both conversions have been heavily promoted and subsidized, their share of primary energy consumption (using the standard Eurostat conversions) was 0.94 percent in 2010 and 1.26 percent in 2011 (Eurostat 2013).

This means that the non-carbon electricity's contribution remains the highest in countries with large shares of hydroelectric or nuclear generation. France has been the global leader: in 2011 fission supplied 77.7 percent of its electricity compared to the global average of 12.3 percent (NEI 2013). After adding the country's hydro generation this advance helped to lower the country's carbon intensity of TPES from 24.5 kilograms of carbon per gigajoule in 1950 to just 10.1 kilograms of carbon per gigajoule in 2000, the rate fell to just below ten kilograms of carbon per gigajoule in 2010 (60 percent drop in six decades). In the US the combination of hydro and nuclear generation lowered the average carbon intensity of TPES from 20.3 kilograms of carbon per gigajoule in 1950 to 15.2 kilograms of carbon per gigajoule by 2010 (25 percent drop in 60 years). Relative decline for the same period was much smaller for the pre-Fukushima Japan (just nine percent) and, despite the country's emergence as the world's largest hydroelectricity producer, little less than 25 percent in China. In global terms the average carbon intensity of TPES fell by 22 percent, from almost 25 kilograms of carbon per gigajoule in 1950 to 19.3 kilograms of carbon per gigajoule in 2010.

What It All Means

The table summarizes long term-declines of average national and global carbon intensities for both total fuel combustion and TPES for the period from 1900 to 2010. Ratios in the last column show that during those eleven decades fuel substitutions lowered the average carbon intensity of fuel combustion by 30 to 35 percent in affluent countries, but by only about 20 percent in China

>

and worldwide, while the carbon intensity of TPES fell by 40 to 50 percent in affluent economies (and about 60 percent in France) but only 25 percent in China and 30 percent worldwide. These realities convey not only the historic pace of the decarbonization process but also suggest the limits to its future extent.

When expressed in terms of the H: C ratio of fossil fuels (leaving out wood), the global indicator of decarbonization rose from 1.0 in 1900 (when coal supplied all but five percent of all fossil energies) to about 1.6 by 1950 and 1.8 by 1980, but during the next two decades it had leveled off at roughly 1.9. By the year 2010 it declined a bit to 1.83. This plateau, followed by a slight decline, has been due to China's extraordinarily large rise in coal extraction, from about 600 megatonnes in 1980 to 3.25 gigatonnes in 2010; that kept China's fossil fuel H: C ratio at 1.15 between 1980 and 2000 and then, even with large increases in consumption of hydrocarbons (crude oil combustion nearly doubling in a decade, natural gas use more than quadrupling) it allowed it to rise only marginally to 1.17 by the year 2010.

When Ausubel (1996) plotted the weighted H:C ratio of global fuel consumption he found that the long-term trends indicated the global mean of about 3.0 by 2010 – but the actual ratio was only 1.83. Ausubel foresaw the arrival of global methane economy (H:C of 4.0) shortly after 2030, and of hydrogen-dominated economy (predicate on massive generation of H_2 without fossil fuels) in the closing decades of the $21^{\rm st}$ and the early decades of the $22^{\rm nd}$ century. Two decades later he still expected, according to the historical trend in decarbonization, "methane to provide perhaps 70% of primary energy soon after the year 2030" (Ausubel 2003, p. 3) and that "large-scale production of carbon-free hydrogen should begin about the year 2020" (p. 6).

In reality, the decarbonization process has slowed down since 1990, the global H: C fuel ratio has been stagnating, and natural gas rose from 26 percent of the world's primary fossil fuel supply to 29 percent by 2000 before declining a bit to 28 percent by 2010. That was a gain of eight percent in two decades – while a gain of 250 percent would be required to lift the gas to 70 percent of the global fossil fuel supply shortly after 2030, clearly a most unlikely shift. Ausubel has been right to foresee a much more important role for natural gas than most people thought possible in the early 1980s - but his anticipated rise of hydrogen, the good element, has been clearly proceeding at a slower pace. Hydraulic fractioning has already boosted production and lower natural gas prices in the US, but few other countries can replicate that pace of expansion: China's vast shale gas deposits are more complex and deeper than those in the US. Moreover, they are located mostly in water-short areas (USEIA 2011), while several European countries with shale gas reserves have already prohibited any recovery of this rich resource.1

Substantial declines in average carbon intensity have not been translated into an absolute decline of global emissions. All Western countries have seen recently a clear moderation in the growth of $\rm CO_2$ emissions: during the 25 years between 1985 and 2010 the US carbon emissions rose by 22 percent – or at most by 24 percent after adding emissions generated by net imports of goods

TABLE: Carbon intensities per gigajoule of fuel combustion and per gigajoule of total primary energy supply (TPES). The table shows the long-term decline, with China and the rest of the world still lagging behind the affluent economies. Data sources for calculating the intensities: Barjot (1991), BP (2012), SB (2013), Schurr and Netschert (1960), Smil (1988, 2010).

| | 1900 | 1950 | 2000 | 2010 | 2010/1900 |
|---------------------------|------|------|------|------|-----------|
| fuel combustion (kg C/GJ) | | | | | |
| France | 26.4 | 24.6 | 17.3 | 18.0 | 0.68 |
| US | 24.7 | 21.0 | 17.8 | 17.8 | 0.72 |
| Japan | 28.3 | 25.2 | 18.9 | 18.6 | 0.65 |
| China | 29.8 | 29.4 | 25.5 | 24.2 | 0.81 |
| world | 27.6 | 25.1 | 23.2 | 21.9 | 0.79 |
| TPES (kg C/GJ) | | | | | |
| France | 26.4 | 24.5 | 10.1 | 9.9 | 0.38 |
| US | 24.1 | 20.3 | 15.6 | 15.2 | 0.63 |
| Japan | 28.0 | 16.7 | 15.4 | 15.2 | 0.54 |
| China | 29.8 | 29.3 | 21.5 | 22.4 | 0.75 |
| world | 27.5 | 24.8 | 21.2 | 19.3 | 0.70 |

produced abroad – compared to 63 percent during the 25 years between 1960 and 1985), those of Japan increased by 18 percent and Germany's emissions had recorded 27 percent decline, largely due to the post-unification collapse of the former East German economy. But these slow-downs and moderate declines have been overwhelmed by large emissions increases in Latin America, Africa and, above all, in Asia: Chinese CO_2 emissions had quadrupled, and Indian releases of fossil carbon rose by nearly 430 percent (CDIAC 2013).

The net result: in 2010 total emissions from combustion of fossil fuels and from all biofuels (wood, charcoal, crop residues, ethanol, biodiesel) were about 10.5 gigatonnes carbon (or 38.5 gigatonnes CO₂), roughly 30 percent higher than in 2000 and 4.2 times above the 1950 level). But further decarbonization gains are certain. Widespread commitment to accelerated transition from coal to natural gas (akin to the recent development in the US) has the potential to push the average global carbon intensity of fuel combustion to 19 kilograms of carbon per gigajoule within two decades, and continued expansion of non-carbon electricity generation could lower the overall carbon intensity of TPES below 17 kilograms of carbon per gigajoule. This means that in the early 2030s specific carbon emissions would be about 20 percent below their 2000 level – but even if the growth of global TPES would average only half of its 1990 to 2010 rate the overall demand would still expand by nearly 25 percent, negating the relative decarbonization gains. This means that only major efficiency gains (greater than those achieved during the past two generations) or an extraordinary commitment to expensive CCS or accelerated transition to non-carbon supplies could bring a slight absolute decline in global carbon emissions.

¹ For detailed and current information on shale gas policies see Shale Gas Europe, Latest News: www.shalegas-europe.eu/en.

The verdict is clear: although the generations-old process of relative decarbonization of the world's fuel supply had experienced a significant slowdown during the second half of the 20th century, reductions in carbon intensity of fuel supply are bound to continue in coming decades, above all thanks to substantial gains in natural gas production; in terms of the overall primary energy supply it should be helped by further gains in hydro, wind and solar electricity. Even so, there are no imminent prospects for any major reductions in absolute emissions of CO₂ – and, of course, the atmospheric behavior and the future levels of absorbed outgoing radiation are determined by the absolute atmospheric levels of the gas, not by reductions of relative burdens per unit of product or service.

RESEARCH

Desulfurization

A Neglected Success Story

Except for some natural gases that are almost pure methane, or mixtures of methane and higher homologs of the straight-chained alkanes series (mostly ethane and propane), all fossil fuels contain some sulfur, with shares ranging from less than 0.5 percent in the lightest (sweet) crude oils to more than four percent in lowquality lignites: 1.5 to two percent is the most common range for bituminous coals and globally traded crude oils (Smil 2008). Fuel combustion generates more than 80 percent of all anthropogenic sulfur with the rest originating largely in the smelting of copper, zinc and lead. Atmospheric oxidations convert the emitted SO₂ to sulfates. The resulting acid precipitation (and also acidifying dry deposition) affects soils, plants and both aquatic and terrestrial ecosystems. It also damages exposed metals as well as limestone and marble (Smil 1997). Beginning in the late 1960s and early 1970s its effects became particularly noticeable across parts of Western and Central Europe, Eastern North America, and China.

The best series of global and regional SO₂ emissions, going back to 1850, and based on emissions inventories and mass balances, were published by Smith et al. (2010) and Klimont et al. (2013). They show total global SO₂ emissions rising from about 20 megatonnes in 1900 to 58 megatonnes by 1950, peaking at just above 130 megatonnes in 1979 to 1980, which means that SO₂ released from fossil fuels (stationary combustion of coal and hydrocarbons, and mobile conversions of liquid fuels on land and water) rose from about 16 megatonnes in 1900 to 105 megatonnes in 1980; their increase was accompanied by rising intensity of emissions, from about 370 grams SO₂ per gigajoule in 1900 to almost 500 grams SO₂ per gigajoule by 1960.

There are three main reasons for the subsequent decline of average sulfur intensity of global energy use: 1. higher shares of hydrocarbons in global energy supply had, on average, much lower sulfur content than coal; 2. high-sulfur crudes and natural gases rich in hydrogen sulfide (H₂S) became routinely desulfurized (in refineries and gas treatment plants) before combustion; 3. after 1980 flue gas desulfurization (FGD) became a common component in large coal-fired power plants, with the adoption starting in the US and Europe, then spreading worldwide. FGD removes SO₂ from exhaust flue gases by reactions with basic compounds (mostly lime or ground limestone); calcium sulfate (CaSO₄), its main product, is either land-filled or it can be used in wallboard manufacture.

By the year 2000 nearly 40 percent of the world's coal-generating capacity had some level of FGD (Smil 2008). Emissions declines were impressive. In the US they were cut nearly by half between 1970 - the first year the United States Environmental Protection Agency (USEPA) began to make consistent calculations and 2000 (USEPA 2013b). In Europe the emissions decreased by half between 1980 and 2004 to 15 megatonnes SO2 (Vestreng et al. 2007). And the average global SO₂ intensity of emissions from fossil fuel combustion was reduced from about 360 grams SO₂ per gigajoule in 1980 to about 150 grams sulfur per gigajoule in the year 2010 when the total emissions from fossil fuel combustion declined to about 82 megatonnes (Smith et al. 2010). But this decline was far from universal: as emissions were cut in Europe and North America, they were rising rapidly in the fast growing Asian economies China and India. Between 1990 and 2005 both China's and India's total SO₂ emissions doubled, respectively to more than 32 megatonnes and nearly ten megatonnes per year: the combination of deep cuts in the West and large increases in the East resulted first in a slight rise of global SO2 releases between 2000 and 2005 and then in a roughly ten percent decline by 2010.

Rising share of Asian emissions has also introduced a greater uncertainty into the global estimates. For example, a comparison of half a dozen estimates of SO₂ emissions by China, the world's worst polluter, shows that the difference between the lowest and the highest value was 38 percent for the year 2000 and 35 percent for the year 2005 with, predictably, the lowest totals claimed by China's Ministry of Electric Power (Lu et al. 2010). Whatever the actual total might be, it seems that even the Chinese emissions finally began to decline after 2007 because of FGD installed at many of China's new (and some older) coal-fired power plants, a reduction that has been confirmed by the Ozone Monitoring Instrument aboard National Aeronautics and Space Administration's (NASA) Aura satellite (Li et al. 2010). Another post-2007 event that helped to reduce the global SO₂ emissions was the worst post-World War II economic crisis. Klimont et al. (2013) showed that by 2011 the global emissions were nearly a third lower than the global record in 1980.

Even accounting for the uncertainties cannot change this conclusion: unlike in the case of CO₂ emissions, global desulfurization has taken place not only in relative but also in absolute terms, we have seen not only lower sulfur intensities per unit of liberated energy but substantial decline in total sulfur emissions. And we have accomplished this despite a massive increase in global combustion of coal - thanks to an effective technical fix (FGD). With continuing transition to natural gas and with almost universal deployment of FGD in coal-fired power plants there is an excellent prospect for further reduction of SO₂ emissions on both national and global scales.

Conclusion: Limits of Decarbonization

Unfortunately, decarbonization of flue gases is a much greater technical challenge than their desulfurization. Recent years have seen too many exaggerated promises regarding the near-term performance of CCS, and too many unrealistic forecasts (Metz et al. 2005, Muradov and Veziroglu 2012, Global CCS Institute 2013). During the next two decades it should become clear if effective (and affordable) decarbonization of large stationary sources is a practical solution or if the best way to decarbonize is to keep changing the make-up of primary energy supply.

On that score we must also remain realistic. The world, contrary to some impressions one might gain by reading European green stories,² is not going to discard fossil fuels any time soon. Our dependence on fossil fuels remains a matter of existential necessity: according to the International Energy Agency (IEA) 81 percent of the world's TPES came from fossil fuels in 2010 (IEA 2012), according to BP (2012) the share was 87 percent in 2011 (the difference is largely due to conversion of primary electricity to the common energy denominator) - while the new renewables (mostly wind and solar electricity) supplied just 1.6 percent of the global TPES. Moreover, indisputable historical evidence (an expression of many technical and infrastructural imperatives) shows that energy transitions are always gradual, protracted affairs and our dependence on fossil fuels will remain high for decades to come (Smil 2010). Hydrogen economy, the ultimate solution to dependence on carbon, remains in the realm of promises and speculations (U.S. House of Representatives 2006, Ball and Wietschel 2010).

But even after another century the historical progression of the H:C ratio may not terminate at pure hydrogen. A less than ideal outcome is more likely because high-density chemical energy will be always preferred for many uses, and liquid hydrocarbons in general, and gasoline and kerosene in particular, are a near-perfect choice, not only due to their high energy densities but also due to their other desirable properties. Decarbonization may thus trend toward optimal carbonization, with a H:C ratio at 4.0 for stationary applications, and close to 2.0 for mobile uses.

Consequently, we will continue to interfere in global biogeochemical cycles of carbon and sulfur. The good news is that the interference in the former is now proceeding with reduced relative intensity and the interference in the latter is, finally, decreasing even in absolute global terms. But if the concerns about the former conform to current expectations (that is, if we will experience rising tropospheric temperatures caused primarily by anthropogenic emissions of greenhouse gases) then the latter news may not be so good because, beyond any doubt, atmospheric sulfates have had a cooling effect, partially counteracting the warming effect of CO_2 , methane and nitrous oxide ($\mathrm{N}_2\mathrm{O}$) (Lelieveld and Heitzenberg 1992, Mendelsohn et al. 2001, IPCC 2007). In any case,

the next two decades will be a critical period: by its end we should have a much better appreciation of the most likely consequences arising from more than a century of human intervention in two key biogeochemical cycles.

References

- Ausubel, J. H. 1996. Can technology spare the Earth? *American Scientist* 415/84: 166–178
- Ausubel, J. H. 2003. Decarbonization: The next 100 years. New York: Rockefeller University. http://phe.rockefeller.edu/AustinDecarbonization.pdf (accessed October 14, 2013).
- Ausubel, J. H., C. Marchetti. 2007. *Wood's H: C ratio*. New York: Rockefeller University. http://phe.rockefeller.edu/docs/WoodsHtoCratio.pdf (accessed October 14, 2013).
- Ball, M., M. Wietschel. 2010. The hydrogen economy: Opportunities and challenges. Cambridge, UK: Cambridge University Press.
- Barjot, D. 1991. L'énergie aux XIXe et XXe siècles. Paris: Presses de l'E.N.S. BP (British Petroleum). 2012. BP statistical review of world energy. London: BP. www.bp.com/assets/bp_internet/globalbp/globalbp_uk_english/reports_ and_publications/statistical_energy_review_2011/STAGING/local_assets/pdf/statistical_review_of_world_energy_full_report_2012.pdf (accessed October 14, 2013).
- CDIAC (Carbon Dioxide Information Analysis Centre). 2013. Fossil-fuel CO₂ emissions. Oak Ridge, TN: CDIAC. http://cdiac.ornl.gov/trends/emis/meth_reg.html (accessed October 14, 2013).
- Cornwell, W. K. et al. 2009. Plant traits and wood fates across the globe: Rotted, burned, or consumed? Global Change Biology 15: 2431–2449.
- Eurostat. 2013. Renewable energy primary production: Biomass, hydro, geothermal, wind and solar energy. http://epp.eurostat.ec.europa.eu/tgm/table.do?tab=table&init=1&plugin=1&language=en&pcode=ten00082 (accessed October 14, 2013).
- Global CCS Institute (Global Carbon Capture and Storage Institute). 2013. Large-scale integrated CCS projects. www.globalccsinstitute.com/projects/browse (accessed October 14, 2013).
- Hester, R. E., R. M. Harrison (Eds.). 2002. *Global environmental change*. London: Royal Society of Chemistry.
- Hey, C. 2012. Low-carbon and energy strategies for the EU The European Commission's roadmaps: A sound agenda for green economy? GAIA 21/1: 43–47.
- Hudson, J. L., G. T. Rochelle (Eds.). 1982. Flue gas desulfurization. Washington, D. C.: American Chemical Society.
- IEA (International Energy Agency). 2012. Key world energy statistics. Paris: IEA. www.iea.org/publications/freepublications/publication/kwes.pdf (accessed October 14, 2013).
- IPCC (Intergovernmental Panel on Climate Change). 1990. *Climate change: The IPCC scientific assessment.* Cambridge, UK: Cambridge University Press.
- IPCC. 2006. 2006 IPCC Guidelines for national greenhouse gas inventories. Volume 2: Energy. Geneva: IPCC.
- IPCC. 2007. IPCC fourth assessment report: Climate change 2007. Cambridge, UK: Cambridge University Press.
- Klimont, Z., S. J. Smith, J. Cofala. 2013. The last decade of global anthropogenic sulfur dioxide: 2000–2011 emissions. *Environmental Research Letters* 8:1–6.
- Lamlom, S. H., R. A. Savidge. 2003. A reassessment of carbon content in wood: Variation within and between 41 North American species. Biomass and Bioenergy 25: 381–388.
- LBNL (Lawrence Berkeley National Laboratory). 2013. *China energy databook*. Berkeley, CA: LBNL.
- Lelieveld, J., J. Heitzenberg. 1992. Sulfate cooling effect on climate through in-cloud oxidation of anthropogenic SO₂. Science 258:117–120.
- Li, C. et al. 2010. Recent large reduction in sulfur dioxide emissions from Chinese power plants observed by the Ozone Monitoring Instrument. *Geophysical Research Letters* 37, doi: 10.1029/2010GL042594.

² For the ongoing discussion of low-carbon and energy strategies for the EU see Hey (2012).

- Lu, Z. et al. 2010. Sulfur dioxide emissions in China and sulfur trends in East Asia since 2000. Atmospheric Chemistry and Physics 10: 6311-6331.
- Mackenzie, F. 2011. Our changing planet: An introduction to earth system science and global environmental change. Upper Saddle River, NJ: Prentice Hall.
- McIlvaine (The McIlvaine Company). 2009. Chinese utility plans. Northfield, IL: The McIlvaine Company.
- Mendelsohn, R., M. Schlesinger, L. Williams. 2001. The climate impacts of sulfate aerosols. Integrated Assessment 2: 111-122.
- Metz, B., O. Davidson, H. de Coninck, M., Loos, L. Meyer (Eds.). 2005. IPCC special report on carbon dioxide capture and storage. Cambridge, UK: Cambridge University Press.
- Muradov, N. Z., T. N. Veziroglu. 2012. Carbon-neutral fuels and energy carriers. Boca Raton, FL: CRC Press.
- NEI (Nuclear Energy Institute). 2013. World statistics: Nuclear energy around the world. www.nei.org/resourcesandstats/nuclear_statistics/worldstatistics (accessed October 14, 2013).
- Nordhaus, W. D. 2008. A question of balance: Weighing the options on global warming policies. New Haven, CT: Yale University Press.
- Rhode, H. 1989. Acidification in a global perspective. Ambio 18: 155-159.
- Royal Ministry of Foreign Affairs. 1971. Air pollution across national boundaries. Stockholm: Royal Ministry of Foreign Affairs.
- SB (Statistics Bureau). 2013. Historical statistics of Japan. Tokyo: SB. www.stat.go.jp/english/data/chouki/index.htm (accessed October 14, 2013).
- Schurr, S. H., B. C. Netschert. 1960. Energy in the American economy 1850-1975. Baltimore, MD: Johns Hopkins University Press.
- Shafizadeh, F. 1981. Basic principles of direct combustion. In: Biomass conversion processes for energy and fuels. Edited by S. S. Sofer, O. R. Zaborsky. New York: Plenum. 103-124.
- Sjöström, E. 1993. Wood chemistry: Fundamentals and applications. San Diego, CA: Academic Press.
- Smil, V. 1988. Energy in China's modernization. Armonk, NY: M. E. Sharpe. Smil, V. 1997. Cycles of life: Civilization and the biosphere. New York:
- Smil, V. 2008. Energy in nature and society: Energetics of complex systems. Cambridge, MA: MIT Press.
- Smil, V. 2010. Energy transitions. Santa Barbara, CA: Praeger.

Scientific American Library.

- Smil, V. 2013. Harvesting the biosphere: What we have taken from nature. Cambridge, MA: MIT Press.
- Smith, S. J. et al. 2010. Anthropogenic sulfur dioxide emissions 1850–2005. Atmospheric Chemistry and Physics Discussions 10: 16111-16151.

- USDA (United States Department of Agriculture). 2010. Wood handbook. Madison, WI: USDA. www.fpl.fs.fed.us/documnts/fplgtr/fpl_gtr190.pdf (accessed October 14, 2013).
- USEIA (United States Energy Information Administration). 2011. World shale gas resources: An initial assessment of 14 regions outside the United States. Washington, D. C.: U.S. Department of Energy. www.adv-res.com/pdf/ ARI EIA Intl Gas Shale APR 2011.pdf (accessed November 6, 2013).
- USEPA (United States Environmental Protection Agency). 2013 a. Flue gas desulfurization system capabilities for coal-fired steam generators. Technical Report, Volume 2. Washington, D.C.: USEPA.
- USEPA. 2013 b. National emissions inventory (NEI) air pollutant emissions trends data. www.epa.gov/ttnchie1/trends (accessed October 14, 2013).
- U.S. House of Representatives (United States. Congress. House. Committee on Science. Subcommittee on Research). 2006. Fueling the future: On the road to the hydrogen economy. Joint hearing before the Subcommittee on Energy and the Subcommittee on Research, Committee on Science, House of Representatives, One Hundred Ninth Congress, first session, July 20, 2005. Serial No. 109-23. Washington, D. C.: U.S. Government Printing Office. www.gpo.gov/fdsys/pkg/CHRG-109hhrg22549/html/ CHRG-109hhrg22549.htm (accessed November 6, 2013).
- Vestreng, V. et al. 2007. Twenty-five years of continuous sulphur dioxide emission reduction in Europe. Atmospheric Chemistry and Physics 7: 3663-3681.
- WNA (World Nuclear Association). 2013. WNA Reactor Database. www.world-nuclear.org/NuclearDatabase/Default.aspx?id=27232 (accessed November 6, 2013).

Submitted May 8, 2013; revised version accepted July 2, 2013.

Vaclav Smil

Born 1943 in Pilsen, former Czech Republic. 1965 doctorate in natural sciences from Carolinum University, Prague. 1972 PhD from Pennsylvania State University, University Park, PA, Faculty of Earth and Mineral Sciences. Distinguished professor emeritus at the University of



Manitoba in Winnipeg, Canada, and Fellow of the Royal Society of Canada. Research interests: interdisciplinary studies of energy, environment, food, population, economy and technical innovation.



Welchen Strom wollen wir haben?

