GLOBAL DISTRIBUTION OF CARBON-DIOXIDE DISCHARGES AND MAJOR ZONES OF SEISMICITY

UNITED STATES DEPARTMENT OF THE INTERIOR
GEOLOGICAL SURVEY
1978
WATER-RESOURCES INVESTIGATIONS 78-39
OPEN-FILE REPORT

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U.S. GEOLOGICAL SURVEY

Water-Resources Investigations 78–39

Open-File Report

May 1978

UNITED STATES DEPARTMENT OF THE INTERIOR

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ABSTRACT

Carbon dioxide discharges of the circum-Pacific belt are in a seismically active zone and in part stem from contact metamorphism. Carbon dioxide discharges in Europe and Asia Minor, also in an area of high seismic activity, are in part from regional metamorphism and are in areas of very high heat flow.

INTRODUCTION

Carbon-dioxide (CO_2)-rich waters have long been of interest. Plinius Secundus (LXXVII, libra XXI) reported on springs at Tungri, which is the present-day Spa, Belgium (Bostock and Riley, 1856), and from springs at Dorylaei (Plinius Secundus, LXXVII, libra V), the present-day Eskisehir, Turkey (Bostock and Riley, 1887).

The production of CO_2-rich waters (commonly also rich in boron) by active metamorphism of carbonate-bearing rocks at depth was first suggested by White (1957). It is evident that water must be driven off because metamorphism at higher temperatures generally is accompanied by dehydration. Many rocks undergoing metamorphism contain carbonates and silicates that may react to release CO_2. Two fluid phases commonly escape under lithostatic pressure gradients, a water-rich liquid (high in bicarbonate, boron, and other constituents) and a CO_2-rich vapor which may be high in hydrocarbon gases and H_2S. CO_2 is likely to be the most notable metamorphic constituent in surface effluents, occurring either as CO_2-rich vapor or as anomalously large amounts of bicarbonate dissolved in water. The associated water is commonly meteoric in origin. We know metamorphic water is seldom discharged at the surface as springs because the metamorphic water is so anomalous in chemical and isotopic composition that it is recognizable even if mixed with meteoric water (White, Barnes, and O'Neil, 1973). The presumed metamorphic water cannot be diluted by meteoric water where pressures are lithostatic, because the meteoric waters would have to flow against the gradient; however, dilution probably is extensive at hydrostatic pressures. Mixing of metamorphic and meteoric waters is suspected where contents of bicarbonate and boron are large even though the isotopic composition is similar to that of meteoric water. Metamorphic water in amounts of less than 10 percent may not be detected isotopically in a mixture of metamorphic water and meteoric water.

The relation of CO_2 discharges to major faults and lithic provinces in California was shown on an earlier map by Barnes, Irwin, and Gibson (1975). Aseismic movement (creep) along certain segments of the San Andreas fault probably is related to structurally diverted, CO_2-charged water (Irwin and Barnes, 1975). The apparent relation between CO_2 discharges and seismicity led to the present global compilation.

For our compilation, a CO_2-rich discharge is defined as that of a gas containing more than 90 percent CO_2 by volume by chemical analysis, or as a water that contains at least 1 gram per liter (g/L) dissolved CO_2 or 1 g/L bicarbonate (HCO_3^{-1}) and with a pH less than 8.3. Adsorption of CO_2 from the atmosphere by alkaline waters may be indicated by a pH greater than 8.3. Specifically excluded from this compilation are formation waters from oil fields. Although some of those formation waters are reported to contain over 1 g/L HCO_3^{-1}, Wiley and others (1975) showed much of the "HCO_3^{-1}" to be acetate (CH_3COO^-) and propionate ($CH_3CH_2COO^-$) ions.

DISTRIBUTION OF CO_2

The CO_2 discharges are concentrated in two major areas of the world (pl. 1). One is a narrow circum-Pacific belt that extends for more than 30,000 km from New Zealand through the island chain of eastern Asia, through western North America to South America. The other major concentration is a broad area in central and southern Europe and Asia Minor. In both areas the geographic coincidence with major zones of historical seismicity is generally striking. Isolated CO_2 discharges also tend to occur in locally seismic areas. Zones of CO_2 occurrences coincide with some zones of ultramafic rocks and blueshist facies rocks. Equally impressive is the general absence or scarcity of CO_2 discharges and seismicity in Australia and in most other major old cratonic regions of the world. The similarity in distribution of CO_2 discharges and zones of seismicity suggests that the production of CO_2 is related to fundamental tectonic processes that are operating widely at the present time. Some CO_2 discharges, however, are near young or active volcanoes, and in regions of high heat flow.

Both the CO_2 discharges and the seismic epicenters are concentrated mainly along or near the boundaries of major crustal plates, and probably result from the interaction of the plates. It is along these active plates that the resistance to plate motion is expressed as seismic energy and anomalous temperatures. Here, conditions are appropriate for release of CO_2 from marine carbonate-bearing rocks through processes of subduction and igneous intrusion. In addition, fractures extending to sufficient depth may also yield CO_2 from the mantle. Within these tectonically active zones, carbon dioxide is being discharged from rocks as old as Precambrian and as young as Holocene. The generation of CO_2 is a longterm event compared to the historical record of seismicity, and thus may be a useful parameter in regional seismic zoning. In old cratonic regions the general absence of both CO_2 discharges and seismicity indicates the continuance of a long history of tectonic quiescence in those regions.

ORIGIN OF CO_2

Worldwide, CO_2 issues from metamorphic rocks as well as from rocks that have no metamorphic history. The CO_2 is thought generally to come from three different sources: (1) organic material, (2) metamorphism of marine carbonate rocks, and (3) the mantle. Stable isotope data are helpful in identifying the sources of CO_2, especially where there is no evident near-surface source. Organic material, in the forms of coal, fossil wood, and petroleum, is rather depleted in ^{13}C (carbon–13) with values of less than -20 permil (Craig, 1953). The ^{13}C isotope compositions of marine carbonates are approximately 0 permil relative to the PDB (Peedee belemnite) standard (Craig, 1953). The most direct evidence for the isotope compositions of CO_2 from mantle sources is provided by inclusions in volcanic rocks erupted along spreading ridges. Moore and others (1977) report ^{13}C compositions from -4.7 to -5.8 permil for fluid inclusions in basalt samples from the Pacific Ocean. Pineau and others (1976) report ^{13}C compositions of -7.6 ± 0.5 permil for CO_2 inclusions in tholeiitic rocks from the mid-Atlantic Ridge. Thus, mantle-derived CO_2 is evidently in or near the range of -4.7 to -8.0 permil.

Even where CO_2 discharges from or near young volcanoes, the stable isotope compositions may be useful to verify the source of the CO_2. Many volcanic deposits rest on marine sediments, and there the source may be volcanic only in the sense that CO_2 originates where the melt originates. CO_2 may also be of metamorphic origin, stemming either from a local high-temperature environment or from direct reaction of silicate melts with sedimentary rocks. The CO_2 from sedimentary rocks may come either from organic material or from carbonate minerals. Still another source of CO_2 may be degassing of the mantle, which may or may not be accompanied by eruptions of mantle-derived silicates. Surficial discharge of CO_2 from complex environments may be derived from a combination of these sources.

An example of the use of isotopic compositions to determine the source of the CO_2 is a study of the carbon dioxide discharging from the Bohemian Massif at Karlovy Vary. There the bedrock is Variscan granite, which is a singularly poor source for either the CO_2 or the calcite that forms extensive travertine deposits. The isotopic composition of the CO_2, near 0 permil, strongly suggests that the source of the CO_2 is marine carbonate-bearing rocks that must underlie the Bohemian Massif (Barnes and O'Neil, 1976).

High thermal gradients tend to coincide with CO_2 discharges, but more than merely high temperature is required to generate CO_2. Simple thermal decomposition of calcite yields lime (CaO), an uncommon mineral. Silica and other oxides are required to form metamorphic calcium minerals. The CO_2 found discharging is a product of a chemical reaction.

Metamorphism to produce CO_2 may occur in a large variety of physical and chemical conditions, and the various conditions will be identified as more detailed studies are made. As an excellent example of the detailed studies needed, Muffler and White (1968, 1969) have described the metamorphic conditions in clastic sediments of the Salton Sea area of California. The sediments are in part detrital calcite and dolomite from marine carbonate rocks of the Colorado Plateau. The calcite and dolomite have $\delta^{13}C$ contents that average ~ -2 permil. Dolomite reacts at temperatures as low as 180°C, and where the produced CO_2 can escape calcite disappears near 300°C. Incomplete decarbonation at depths below 1000 m and temperatures above 250°C (Clayton and others, 1968) tend to have $\delta^{13}C$ contents from -3 to -5 permil, suggesting that ^{13}C is selectively concentrated in escaping fluids.

There are only two requirements for a chemical reaction to take place, a nonzero chemical potential and a reaction path or mechanism. As long as a chemical potential and reaction path exist for calcite to react to form another calcic mineral and CO_2, the reaction will occur. The reaction may be localized as in contact metamorphism or dispersed as in regional metamorphism.

Craig (1963) reviewed the complexities and uncertainties of interpretation of ^{13}C data in geothermal areas. In the absence of other information, ^{13}C compositions near 0 permil should be regarded conservatively as being from a marine carbonate source. Fractionations during solution and exsolution added to the range of marine carbonate ^{13}C compositions can probably yield a range in $^{13}CO_2$ isotopic compositions from -5 to $+5$ permil.

The production of a separate CO_2 phase in th rocks may have structural implications. As Lockner and Byerlee (1977) have shown experimentally, high pore pressures markedly reduce the strengths of rocks, and if the least principle stress is vertical, thrust faulting may occur. Hubbert and Rubey (1959) pointed out the role high fluid pressures play in thrust faulting. The importance of high fluid pressures in the tectonics of the California Coast Ranges was described by Berry (1973). Although these authors considered water alone, CO_2 as a separate phase would be just as effective in forming and transmitting high fluid pressure.

MAP COMPILATION

The chemical criteria for the CO_2 discharges are arbitrary but during the course of this compilation it became obvious that changing the concentration limits to values as low as 0.5 g/L (still an unusually high HCO_3^- concentration) would serve no purpose other than to provide more occurrences from the same general areas already established and would not change the general pattern of occurrences. The sources of data for the CO_2 discharges are given separately from the references cited in the text. For uniformity all sources of data have been translated into English. A first map compilation on a scale of approximately 1:40,000,000 is inevitably incomplete. Unfortunately, no data were found for CO_2 discharges in the People's Republic of China, although reportedly such occurrences exist (Šilar, 1961). The authors would appreciate additional information on the location and the chemical and isotopic compositions of CO_2-rich waters.

The CO_2 occurrences and the major zones of seismicity are compiled on a base map that shows the global distribution of alpine-type ultramafic rocks and blueschists (Irwin and Coleman, 1972). Additional blueschist localities are from Dobretsov (written commun., 1973). It should be noted that the zones of seismicity are generalized from Tarr (1974), with the exception of data from Fairhead and Henderson (1977) for Africa, Lee and others (1978) for China, and NOAA (1970) for the north polar region. Tarr's (1974) map shows only magnitude 5 and greater earthquakes and is restricted to the period 1963–72. A more inclusive and lengthy record of seismicity would show an even closer correlation with the CO_2 discharges.

CIRCUM-PACIFIC BELT

A roughly continous belt of CO_2 discharges, more than 30,000 km long, stretches from southern South America through Central America, western North America, Kamchatka, Japan, Taiwan, and New Guinea to New Zealand along the circum-Pacific belt.

In the Matsushiro area of Japan, CO_2 discharges associated with earthquakes have been documented by Yoshioka and others (1970). In the same area, Wakita and others (written commun., 1977) reported $\delta^{13}C$ values from −1 to −3 permil and interpreted the source of the CO_2 to be Paleozoic marine carbonates. Wakita and others attributed the high value of 8.9×10^{-6} for the $^3He/^4He$ ratio for the He in the CO_2 to a mantle-derived source. They interpret the He to be derived from a small intrusion into the crust accompanied by reactions with marine carbonates to yield CO_2.

On the Alaska Peninsula at Gas Rocks (57°51′40″N, 156°30′00″W) an explosive volcanic episode began in April 1977 (T. P. Miller, U.S. Geol. Survey, oral commun., 1977). In August 1977 the copious gas discharge near Gas Rocks was sampled by G. A. McCoy, U.S. Geological Survey. The gas was analyzed for chemical and isotopic compositions in the U.S. Geological Survey laboratories in Menlo Park, California. The gas is 98 percent CO_2 and its ^{13}C composition is −6.36 permil. The content of ^{13}C is too low for a marine carbonate source but is in the range of values for CO_2 from the mantle. The explosive eruption at Gas Rocks is probably due to an abrupt release of CO_2 because the pressure of CO_2 dissolved in the melt exceeded the total pressure.

The broad belt of CO_2 occurrences of the western United States may be subdivided into three subsidiary belts: (1) a coastal (Franciscan Formation) geochemical province, (2) Sierra Nevada, and (3) Rocky Mountains.

Franciscan rocks of the coastal geochemical province discharge carbon dioxide (Barnes, 1970), usually accompanied by methane (Barnes and others, 1973). A current study shows that CO_2 from Franciscan rocks ranges from +5.2 to −14.0 permil. From the data at hand there is no relation between the ^{13}C data and existence of travertine deposits or the presence of methane in the gases. The CO_2 is apparently generated both by the breakdown of organic material depleted in ^{13}C and from solution or metamorphism of calcite of marine origin enriched in ^{13}C. If the mantle also contributes to the CO_2 flux, it is masked by mixtures of CO_2 from other sources. The present phase of metamorphism of the Mesozoic eugeosynclinal sediments of the Coast Range of California is probably in its early geologic history, even though the rocks were previously metamorphosed. Labile organic materials are still present and decomposing. The metamorphism is also accompanied by quite high heat flow.

The Sierra Nevada CO_2 discharges have been studied by Mariner and Presser (written commun, 1978), who report ^{13}C values of −6 to −10 permil. From chemical and isotopic evidence, the fluids are meteoric water and mantle CO_2. The CO_2 must, as a consequence, be present as a separate phase from the mantle to at least as shallow a depth as the meteoric water circulates. To be present as a separate phase the CO_2 pressure must at least equal total (confining) pressure.

In the Rocky Mountains province in Colorado, CO_2 species in CO_2-rich springs have been analyzed for ^{13}C (U.S. Geol. Survey, unpub. data, 1977). The ^{13}C-enriched CO_2 (−1.96 permil) found at Rico may be from the metamorphism of shaly limestone and limy shale that were described by McKnight (1974), or from deeper sources. Impure marbles of a Precambrian metamorphic complex occur near Guffey (D.S. Sheridan, oral commun., 1977). The $\delta^{13}C$ value of −1.45 permil in the CO_2 discharging near Guffey may be from the metamorphism of the marble. CO_2 discharges from the Precambrian Pikes Peak Granite at Manitou Springs (−3.30 to −2.8 permil) possibly are from a mantle source. If so, mantle-derived CO_2 may be more enriched in $^{13}CO_2$ than indicated earlier. ^{13}C data from complex igneous and metamorphic terranes cannot be unequivocally interpreted without detailed geologic mapping and consideration of multiple origins.

In Yellowstone National Park, Wyoming, USA, in the Rocky Mountains province, CO_2 discharges coincide with extraordinarily high heat flow (Fournier and others, 1976), however the coincidence is not perfect. For example, heat flow in excess of 2 HFU, or at least 30 percent above the world's average conductive heat flow, characterizes most of the broad central and eastern part of the U.S. seismic belt of figure 1 (Diment and others, 1975), but CO_2 occurences are no more abundant than in adjacent areas to the east and north. Conductive heat flow is slow relative to fluid flow. Thus a thermal anomaly at depth may be a source generating CO_2 that discharges from the surface before an increase in heat flux at the surface is noted.

Craig (1961) reports analyses on the CO_2 from the Yellowstone National Park that range from about −1 to −6 permil and averages (13 samples) −2.8 permil. Thus, although the average isotope composition is rather close to average marine limestone, considerable volcanic or mantle CO_2 may be present. Friedman (1970) studied travertines depositing in Yellowstone National Park, and reported that fractionations between dissolved carbon species and travertine range from near equilibrium (4 permil) at 75°C to only 1 permil at 25°C. Neither Craig nor Friedman, however, measured the isotope fractionation between dissolved CO_2 species and CO_2(gas). In the Sierra Nevada and Klamath Mountains of California, USA, Mariner and Presser (written commun., 1977) studied the fractionation between dissolved CO_2 species and CO_2(gas) from the springs. They found that fractionations range from equilibrium (4 permil) down to no fractionation (0 permil). These relatively small fractionations permit identification of sources of the Yellowstone CO_2 gases to be chiefly limestones (Craig, 1953), and allowed Friedman (1970) to conclude that dissolved CO_2 species and travertines of Yellowstone National Park are derived from marine limestone by decarbonation. Leeman and others (1977) supported the limestone source for the travertines from the similarity of lead isotopes in the limestone and travertines.

In summary, the scattered CO_2 discharges of the circum-Pacific belt seem generally to be from metamorphism of carbonate-bearing rocks and from mantle sources. Some widely separated occurrences may be related to isolated intrusions causing contact metamorphism.

EUROPE AND ASIA MINOR

The CO_2 that is discharging from Europe and Asia Minor is derived in large part from metamorphism of marine carbonates, whether from originally impure limestone or from calcareous siliceous sediments. A marine carbonate origin for the CO_2 of the mineral waters of Czechoslovakia was suggested by Barnes and O'Neil (1976) on the ground that the $\delta^{13}C$ values of the high CO_2 waters are close to 0 permil. All the waters discharging with CO_2 in Czechoslovakia are largely or entirely of meteoric origin. Deuterium and $^{18}0$ compositions of the mineral waters of Czechoslovakia (Barnes and O'Neil, 1976) are in excellent agreement with the stable isotope relations of meteoric water (Craig, 1961). The isotopic results confirm the conclusions of Kačura and others (1969) based on hydrogeologic analysis. Dowgiallo and others (1973, 1975) also showed that much of the CO_2 of southern Poland is also from marine carbonates, although ^{13}C-depleted CO_2 is probably from oxidation of organic matter. The ^{13}C data of Buachidze and Buachidze (1976) are near 0 permil and show that the CO_2 of the Caucasus is probably also derived from marine carbonates. Cornides and Kecskés (1974) concluded from ^{13}C data that the CO_2 of Hungarian mineral waters was derived partly from metamorphism of limestone and partly from volcanic sources. By measurements of the isotopic compositions of French mineral springs Risler (1978) showed that the water is of meteoric origin but the CO_2 is largely of igneous (mantle?) origin. In a thorough study of the isotopic compositions of CO_2 in central and southern Italy, Panichi and Tongiorgi (1976) showed that most of the CO_2 is from metamorphism of marine carbonates. Their isotopic data agree with the results separately reported by Manfra, Masi, and Turi (1974). Craig (1963) showed that the waters from Lardarello, Italy, are also chiefly of meteoric origin, but with enrichments in $^{18}0$ due to reaction (exchange) with the rocks. The lack of identified metamorphic or magmatic water accompanying the CO_2 is puzzling. Possibly water that was present earlier either has escaped or has been incorporated in hydrous minerals. Conceivably metamorphic water doesn't reach the surface because after it condenses it is no longer as buoyant as CO_2 gas; alternatively, it may escape but be diluted beyond recognition by meteoric water. The observations of Taylor (1974) and Margaritz and Taylor (1976) of large scale exchange of $^{18}0$ between igneous rocks and meteoric waters is consistent with the failure to find metamorphic water in Europe or Asia Minor.

In Europe, the belts of CO_2 discharge correlate approximately with high heat flow, not only in the Pannonian Basin of Central Europe but also both east and west. The high heat flow in the Caucasus and the Crimea (Petkov and others, 1976) corresponds quite well with the occurrence of CO_2 discharges. To account for the high heat flow of the Pannonian Basin, Stegena and others (1975) advocate mantle-derived intrusion into the overlying sediments. They state that the high heat flow cannot be supported by thermal conduction. Lachenbruch and others (1976) point out that intrusion is not as effective in transferring heat as convective flow in a melt. However, once the melt stops convecting, heat flow decreases until a more normal thermal gradient is established. Convective heat flow and the long subsequent time necessary for recovery may explain why older metamorphic terranes such as the Appalachians and the Urals show so little evidence of present-day metamorphism. Saratoga Springs, New York, is the only discharge of CO_2 known in the Appalachians and none is known in the Urals.

Further evidence for metamorphic reactions in the Roman province is supplied by Hurley and others (1966). On the basis of the $^{87}Sr/^{86}Sr$ ratios and the Rb and Sr contents of potash-rich lavas, they conclude that the lavas are derived from the melting of old sialic crustal rocks. Turi and Taylor (1976) concluded from ^{18}O compositions that the volcanics of the Roman province have undergone moderate to extensive reactions with crustal material, but are at least in part derived from the mantle.

CONCLUSIONS

Regional metamorphism, perhaps in early stages, is probably occurring now in California in eugeosynclinal rocks with their attendant ultramafic rocks. Regional metamorphism, perhaps of a more advanced stage, is occurring in Europe and Asia Minor where the CO_2 production is principally from the breakdown of calcite of a marine origin. Older terranes such as the Appalachian geosyncline may be products of metamorphism such as is now occurring in Europe and Asia Minor.

Relatively isolated metamorphism may be occurring within a belt from southern South America to New Zealand, and is perhaps related to contact metamorphism associated with intrusions. The areal extent of CO_2 production in Europe and Asia Minor is much greater than that in any area found along the circum-Pacific belt with the possible exception of the western United States. Judging from the CO_2 occurrences, if any areas of the world are undergoing regional metamorphism at the present time, they are the orogenic belts of Europe and Asia Minor and Coast Ranges of California. Many of the rocks that are being metamorphosed have already undergone metamorphism one or more times.

ACKNOWLEDGMENTS

We wish to thank our colleagues of the Czechoslovak Geological Survey, Prague, and the Dionyza Stura Geological Institute, Bratislava, for their generous assistance in sampling mineral waters in Bohemia and Slovakia, respectively. We thank the Director General of the Mineral Research and Exploration Institute, Ankara, for chemical data on Turkish mineral waters. We thank N. L. Dobretsov for data on additional blueschist localities in the U.S.S.R.

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