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# Physical and Geological Implications in Surface Geochemical Exploration\*

by  
DIRK KETTEL\*\*

11 figures

## *Abstract*

Results from worldwide surface geochemical surveys published or unpublished as well as from own survey experience have been put together and examined under the scope of their evidence on physical processes detected in laboratory and in situ experiments.

This work has resulted in a new data acquisition and interpretation concept, which actually converges with all new data input. Thus it links up laboratory experience, theoretical considerations and data measured in nature. It obviously fits the processes going on in gas generation and migration sufficiently well as to be applicable as a risk reducing tool in hydrocarbon exploration.

## *Zusammenfassung*

Publizierte und unveröffentlichte, weltweite geochemische Oberflächenmessungen sowie eigene Erfahrungen wurden zusammengestellt und der Rahmen ihrer Übereinstimmung mit experimentell im Labor sowie «in situ» entdeckten physikalischen Prozessen untersucht.

Das Resultat dieser Arbeit besteht in einem neuem Feldarbeit- und Interpretationskonzept welches tatsächlich mit allen neu eingegebenen Daten übereinstimmt. So werden Laborerfahrung, Theorie und gemessene Felddaten miteinander verbunden. Offensichtlich kann damit der Prozess der Gasgeneration und -Migration genügend gut erklärt werden, um als Risiko - reduzierendes Mittel bei der Kohlenwasserstoff-Exploration zu dienen.

## **1. The physical process of gas migration**

Gas data from four shallow wells in the Münsterland basin sampled at intervals of approx. 1 m (KETTEL et al., in preparation) reveal the feature of a pre-steady state migration front wandering through space and time, starting from the top of a gas-saturated Upper Cretaceous section and penetrating into primarily gas-free glacial drift sediments. The starting time of the process is  $t_0 = 12 \times 10^3$  ybp and the migration distance during this time is 0,4 m.

In order to describe the aging physical process this implies, that the gas concentration and isotopic profiles could only be matched applying the model of adsorption chromatography, where a mobile or carrier medium acts as the transporting agent. This results in a diffusion equation with a convectational term.

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The local hydraulic situation implies that a water flux in upward direction could be identified as the carrier medium or solvent. The solute (gas) leaves somewhat behind the solvent front due to adsorption — desorption processes at the surface of a solid adsorbent (clay).

### *Conclusions*

The study cited here shows, that convection, somewhat modified by diffusion in solution, could be identified as the main physical process governing gas migration within the ppb-range in near-surface sediments.

## **2. Starting point: The original source rock or accumulation signature**

The aim of surface geochemical exploration is to realize the existence and properties of source rocks and hydrocarbon accumulations in deeper parts of a basin from surface gas sampling.

The data transmitted to the surface are the molecular and isotopic composition of the gases, where the near-surface data have to be distinguished from the original source rock — or accumulation data.

As shown by FUEX 1980, the isotopic fractionation of gases migrating between the source rock and an accumulation will nearly always be negligible due to the high gas quantities wandering.

The opposite is the case on the way from the source rock or the accumulation towards the surface. The compositional shifts occurring here will be described in the next chapter.

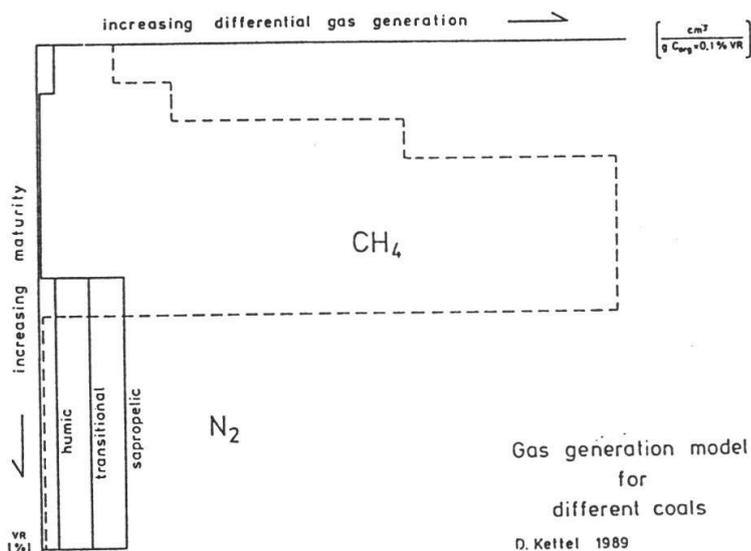
The first condition to be fulfilled when predicting subsurface geology from surface gases is, that the original gas composition from source rocks or in accumulations reflects actual source rock properties such as facies and maturity.

Such correlations are shown in Figs. 2 and 3 for  $\delta^{13}\text{C}_1$  and  $\delta\text{DC}_1$  respectively, where only the  $\delta^{13}\text{C}_1$ -function for gases from marine sources (FABER 1987) was well established until now. The characteristics for more terrestrial facies types are newly reported here.

As the most appropriate case history data set for calibrating gas compositions on more humic facies properties is from the NW and Central European Upper Carboniferous basin, it is utilized here. But, as the relevant source rock thicknesses in this basin vary between some 100 m and approx. 3 000 m, no discrete maturity level can be related to the composition of an overlying hydrocarbon accumulation.

### *The hydrocarbon generation model*

In order to attribute to each case history its individual source rock maturity responsible for the hydrocarbon fill, an effective maturity has been calculated, using the model for differential gas generation with maturity for different humic facies types shown in Fig. 1, the local source rock thickness, the local maturity gradient and the present maturity at the top of the source rock section.



**Fig. 1**  
Differential gas generation  
for different coals

### The $\delta^{13}\text{C}_1$ characteristic

When plotting the  $\delta^{13}\text{C}_1$  from gas accumulations against the respective effective maturities previously described (Fig. 2), they show a good correlation for each facies type. The grouping into facies types can be mapped within the Upper Carboniferous basin (KETTTEL 1989), showing more marine influence in the northwestern part of the basin.

The function for «mixed coal» is in agreement with data from integral pyrolysis by CHUNG and SACKETT 1979 on a coal of the same facies, transferred into differential coalification steps.

### The $\delta\text{DC}_1$ characteristic

The  $\delta\text{DC}_1$  function with increasing effective maturity (Fig. 3) shows also an isotopic fractionation towards the more positive for each facies type, which coincides with pyrolysis measurements from CHUNG 1976 and SACKETT 1978.

In contrast to  $\delta^{13}\text{C}_1$ , the  $\delta\text{DC}_1$  value is more sensible to facies differences at lower maturity levels, where gases from marine source rocks are more positive than gases from more humic sources. This has also been seen by SCHOELL 1984 a. At higher maturities the facies functions coincide more or less.

### Conclusions

Maturity / isotope value correlations have newly been established, which for the first time are in good agreement with the actually valid hydrocarbon generation model. Thus, maturity intervals with a high hydrocarbon generation potential coincide with intervals of a high sensibility of the respective  $\delta$ -values.

## 3. Compositional shifts during migration

As shown in chapter 1, the physical process mainly responsible for gas migration in near-surface sediments could be identified as migration in solution, somewhat retained by adsorption — desorption processes at the surfaces of the clayey particles of

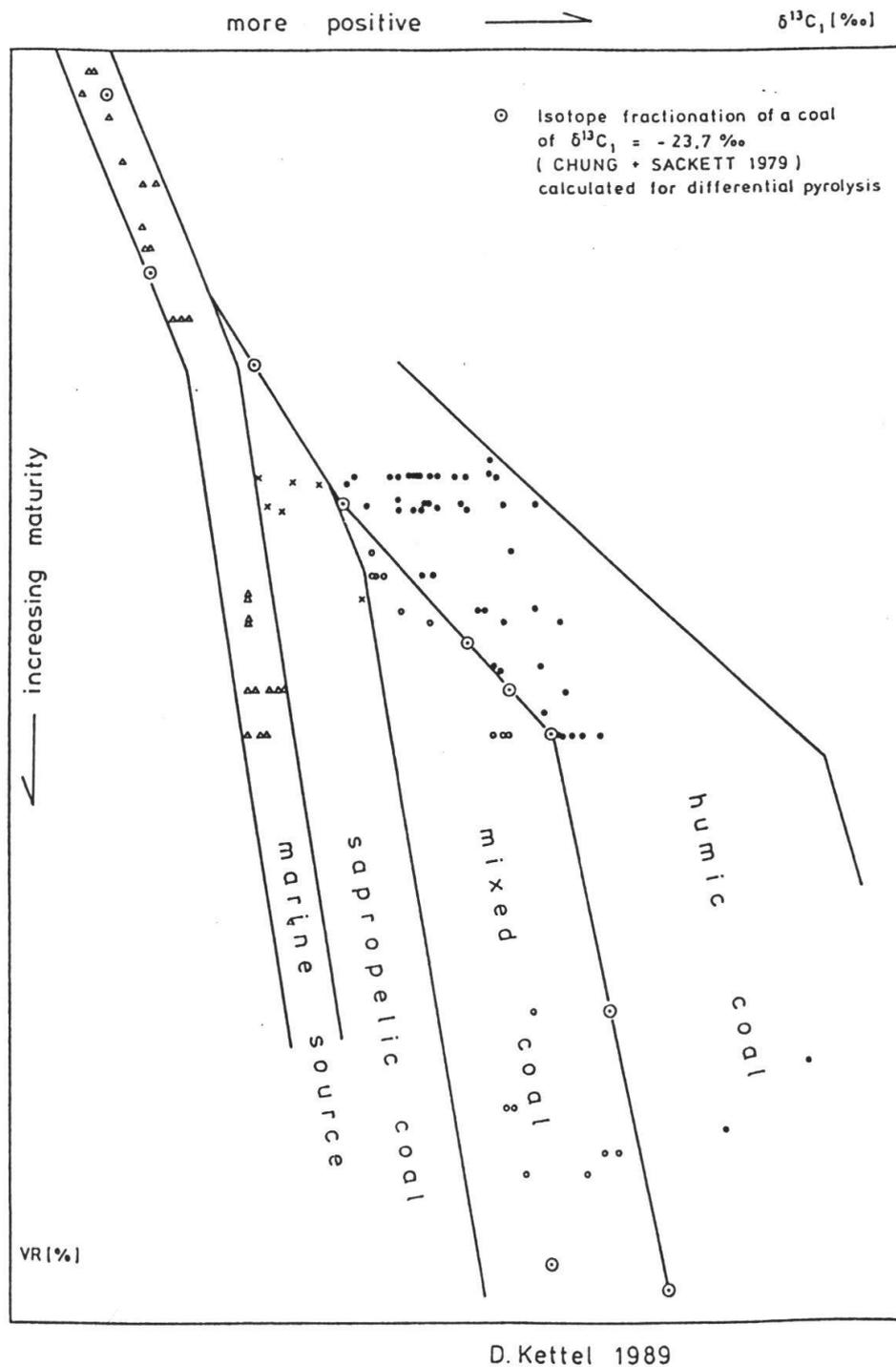


Fig. 2  $\delta^{13}C_1$  - maturity plot

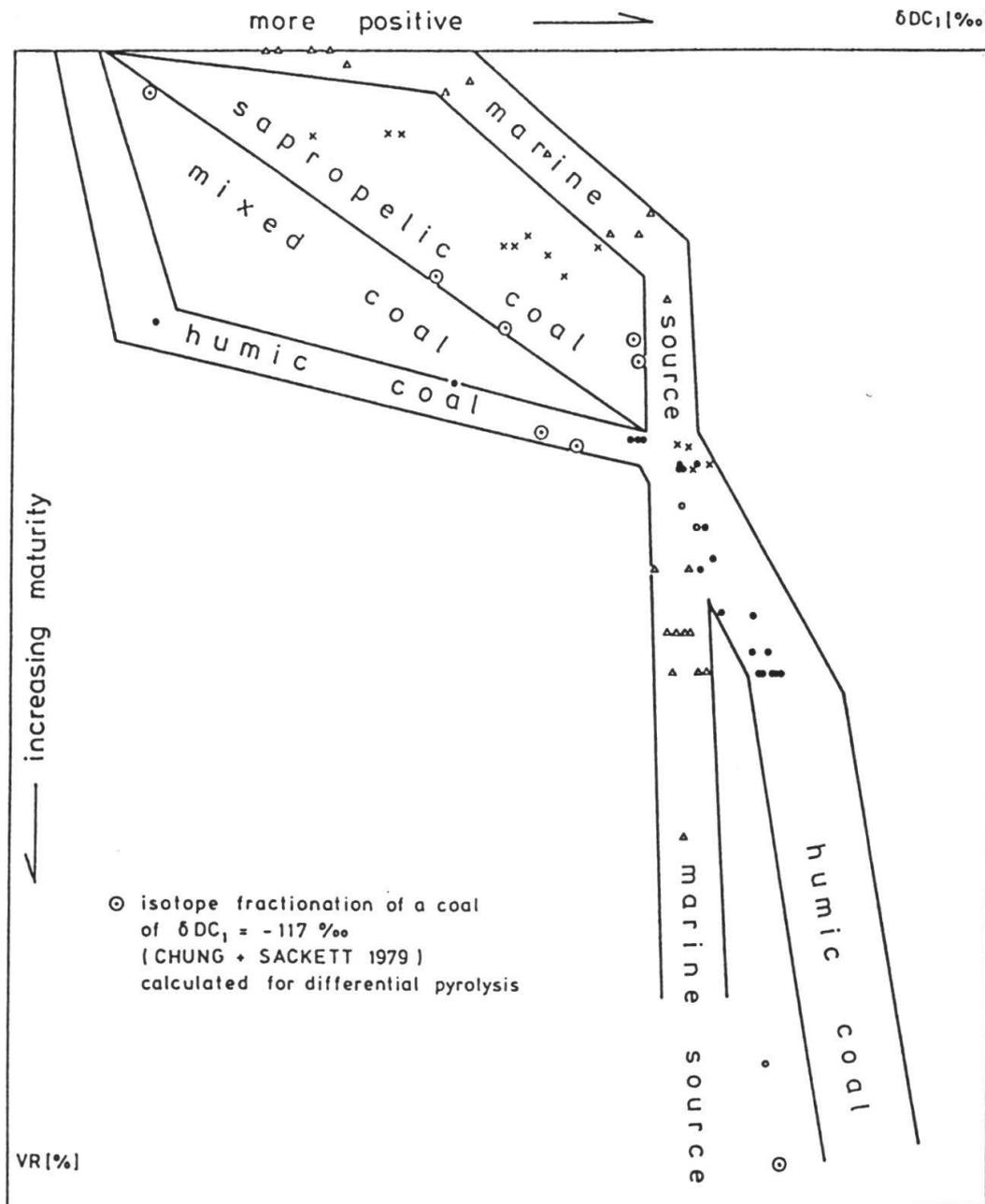
the sediments. So it is dealing with a three-component system: gas — water — solid adsorbent. In surface geochemical exploration we are measuring gases adsorbed at the clayey fraction  $< 63 \mu$ .

From a theoretical point of view, the isotopic fractionation to be expected along a migration path could be calculated using mass balances for the lighter and the heavier isotopes, the definitions of the  $\delta$ -values and an appropriate equilibrium fractionation factor  $\alpha$  for the transition gas in solution — adsorbed gas.

In the present case,  $\alpha$  is derived from geological case histories (wells), whose data have been matched closely by the model.

The basic equation of the model links together the concentration of the adsorbed methane, the isotopic shift and a factor A. Factor A is dependent on several geological parameters (Fig. 9), the most important of which are the permeability of the sampled sediment and the original gas quantity available for migration.

As shown in Figs. 4 and 5, with the same factor A the degree of isotopic shift increases with decreasing gas concentrations. That means, that the real surface expres-



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Fig. 3 δDC<sub>1</sub> - maturity plot

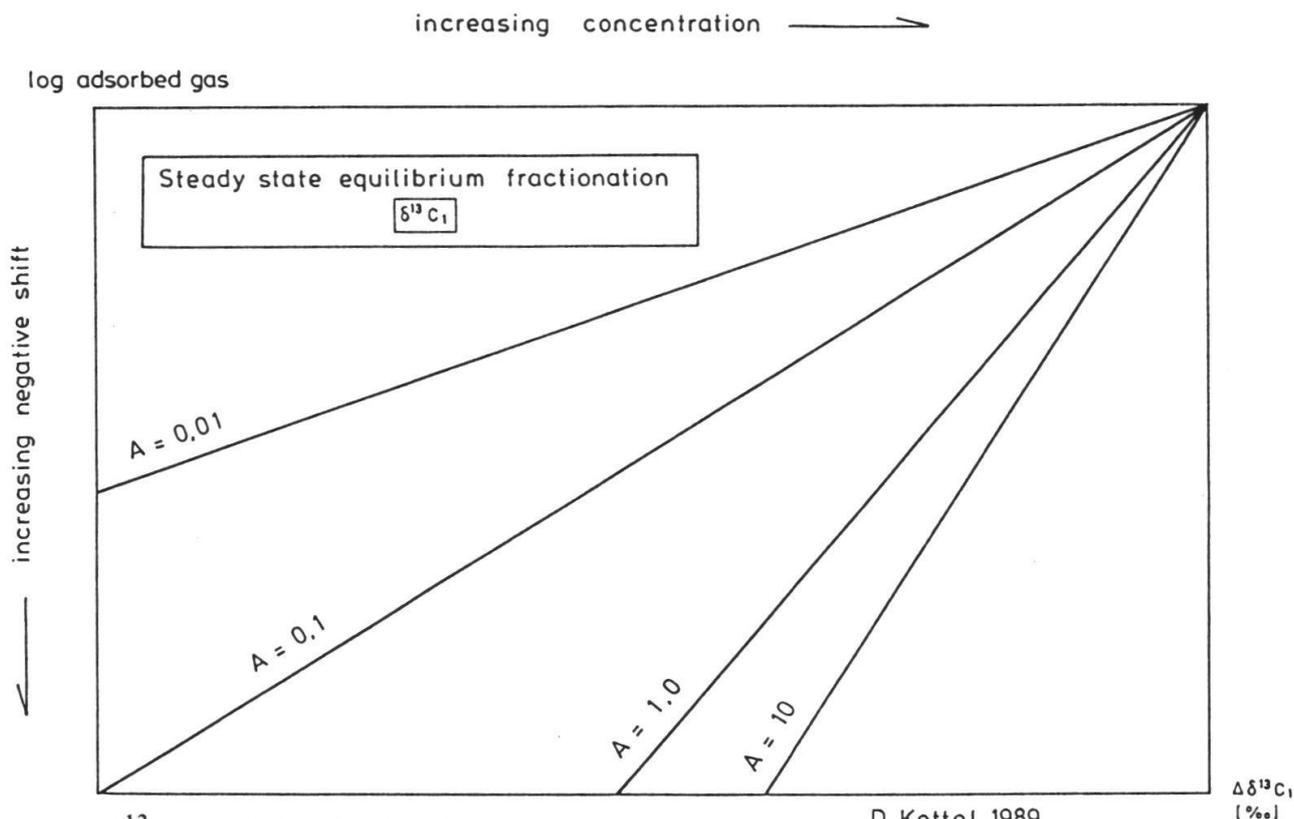


Fig. 4  $\delta^{13}\text{C}_1$  equilibrium fractionation

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$\Delta\delta^{13}\text{C}_1$   
[‰]

sion of a higher gas input from below (accumulation in the subsurface) are not the measured gas concentrations, but are changes in the factor  $A$ , when sample lithology and temperature, water flux and migration time are constant (Fig. 10).

As in geological environments each fractionation process has reached its equilibrium, the  $\delta^{13}\text{C}_1$  (Fig. 4) suffers exclusively isotopic shifts towards the more negative, the  $\delta\text{DC}_1$  (Fig. 5) exclusively towards the more positive, which is in accordance with laboratory experience for the system gas in solution — free gas (FUEx 1980). Also the high  $\Delta\delta\text{DC}_1 / \Delta\delta^{13}\text{C}_1$  value of approximately 18 is in good agreement with results previously reported (GUNTER and GLEASON 1971, GANT and YANG 1964, FUEx 1980).

### Conclusions

HC gas migrating through water-wet pores suffers isotopic and other compositional changes, which can alter the original (source rock or accumulation) signature severely. But as these changes are mainly dependent on the lithology of the sample and on the local gas supply from below, they can be used in determining the original source rock signature and/or possible hydrocarbon accumulations in the subsurface in various basinal settings (see next chapters).

As far as heavier gases are concerned ( $\text{C}_2 +$ ), case histories suggest that no remarkable isotopic fractionation is occurring here during migration. This could be explained by the much higher adsorption probabilities for  $\text{C}_2 +$  than for methane. In surface geochemical exploration this has the advantage, that the  $\text{C}_2 +$  yield is a direct function of the hydrocarbon supply, and can therefore realistically be used in the detec-

tion of hydrocarbon accumulations in some selected basinal settings (for example SANDY, in press). It has the disadvantage that as  $\delta^{13}\text{C}_2$  and  $\delta^{13}\text{C}_3$  do not identify source rock properties nearly as well as the pair  $\delta^{13}\text{C}_1 / \delta\text{DC}_1$  of methane, only poor information on subsurface geology is given by the isotope values of heavier gases.

#### 4. The interpretation of surface data

As could be seen in the previous chapter, in the identification of the original source rock signature and/or of possible changes in the hydrocarbon supply the knowledge of factor A is of primary importance.

Fig. 6 shows a differential diagnosis interpretation scheme of the pair  $\delta^{13}\text{C}_1 / \delta\text{DC}_1$  in order to identify the original source rock signature, which is essentially a composite of Figs. 2, 3, 4, 5. The maturity / facies spurs at the left of the figure represent  $\delta^{13}\text{C}_1 / \delta\text{DC}_1$  pairs of original source rock signatures possible in nature. All isotopic shifts due to migration occur in right hand upward direction (more negative  $\delta^{13}\text{C}_1$ , more positive  $\delta\text{DC}_1$ ) as indicated by the parallel lines. The distance of shift along these lines is a function of factor A and the measured gas concentration. In other words, any measured near-surface pair of  $\delta^{13}\text{C}_1 / \delta\text{DC}_1$  can be shifted back parallel to the lines towards its original source rock pair. If more than one original source rock pair could be involved — which is normally the case at lower maturity levels — the knowledge of factor A will determine the distance of back shifting, and thus discriminate one source rock option.

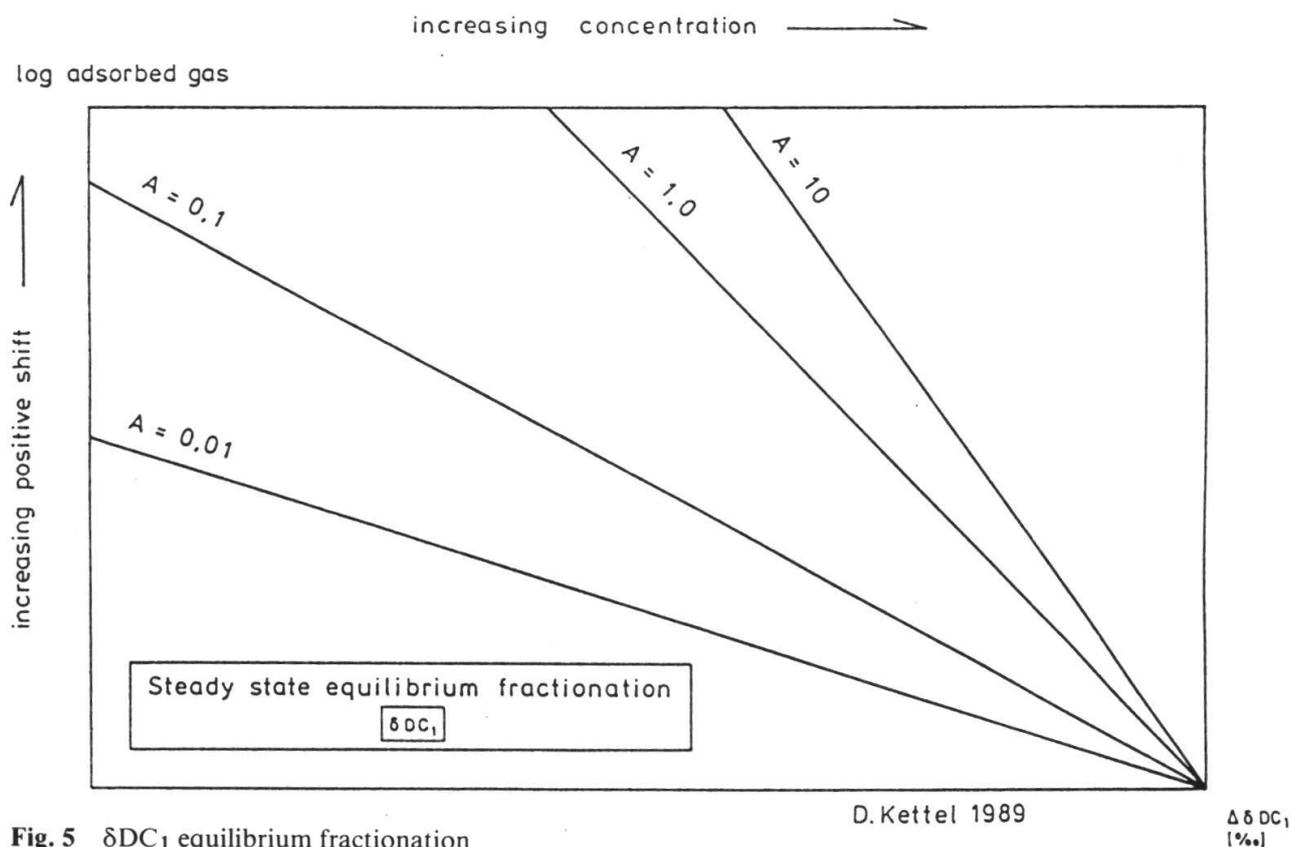


Fig. 5  $\delta\text{DC}_1$  equilibrium fractionation

The determination of factor A could be done in different ways (Fig. 11):

- by determining the basic geological factors responsible for A listed in Fig. 9 from the local and basinal geology, where the permeability of the sample is the most outstanding parameter. Accuracy is here generally not better than one decade.
- by plotting the measured isotope values against the respective yields for samples of the same lithology and the same source rock signature. Any resulting function could be fitted on the graphs Figs. 4 and 5, and thus factor A could be identified. Accuracy could here be in the range of the laboratory reproducibility.
- by plotting the measured isotope values as a function of sample lithologies for samples of the same yield and the same source rock signature. Accuracy could here also be in the order of the laboratory measurements.

All these independent determination procedures must lastly converge on an unambiguous source rock identification.

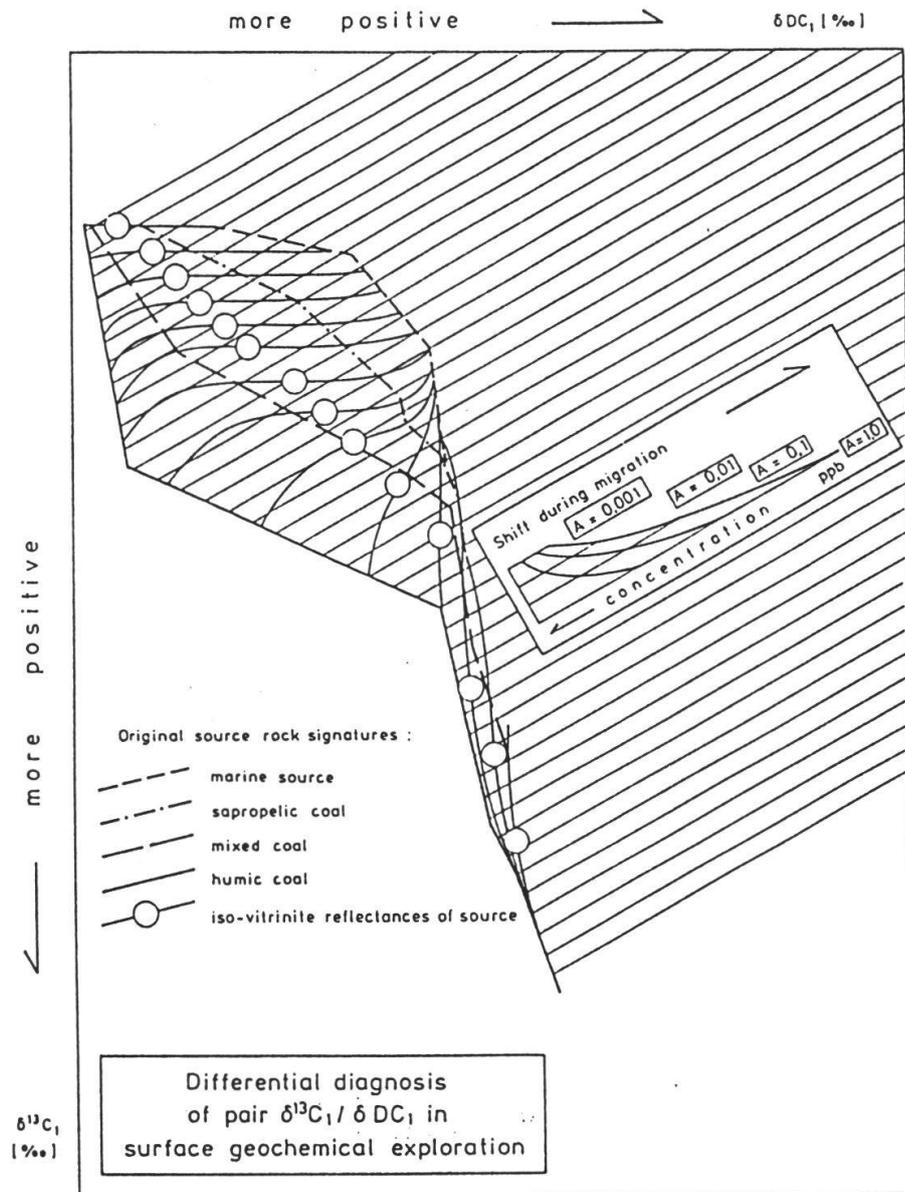


Fig. 6 Differential diagnosis of pair  $\delta^{13}C_1 / \delta DC_1$

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As far as the detection of possible hydrocarbon accumulations is concerned, mapping of factor A could provide a tool for estimating variations in the primary hydrocarbon potential in the subsurface, when other parameters — as indicated in Fig. 10 — are constant, and in special geological settings (see next chapter).

### Conclusions

In the identification procedure of the original source rock properties as well as of variations of the hydrocarbon potential in the subsurface, the determination of factor A, which is essentially a geologic factor, plays an important role.

## 5. The application to different basinal settings

In the pre-evaluation, if a discrete geological setting of a basin is suitable for the application of surface geochemical exploration, the migration velocity of gases and the migration state (steady / non steady) are crucial.

Figs. 7 and 8 show the two main types of an actual basinal status which could be encountered: a basin which underwent young uplift or inversion (Fig. 7), and a steadily subsiding basin (Fig. 8). The configuration of gas parameters in the near-surface layers depends directly on the relationship sedimentation rate / compaction water flow, both in [m/a]. Compaction water flow stands here for gas migration velocity, because a water-transported gas front leaves behind the water front only by the factor of the adsorption probability (KETTEL et al., in preparation), which is very small.

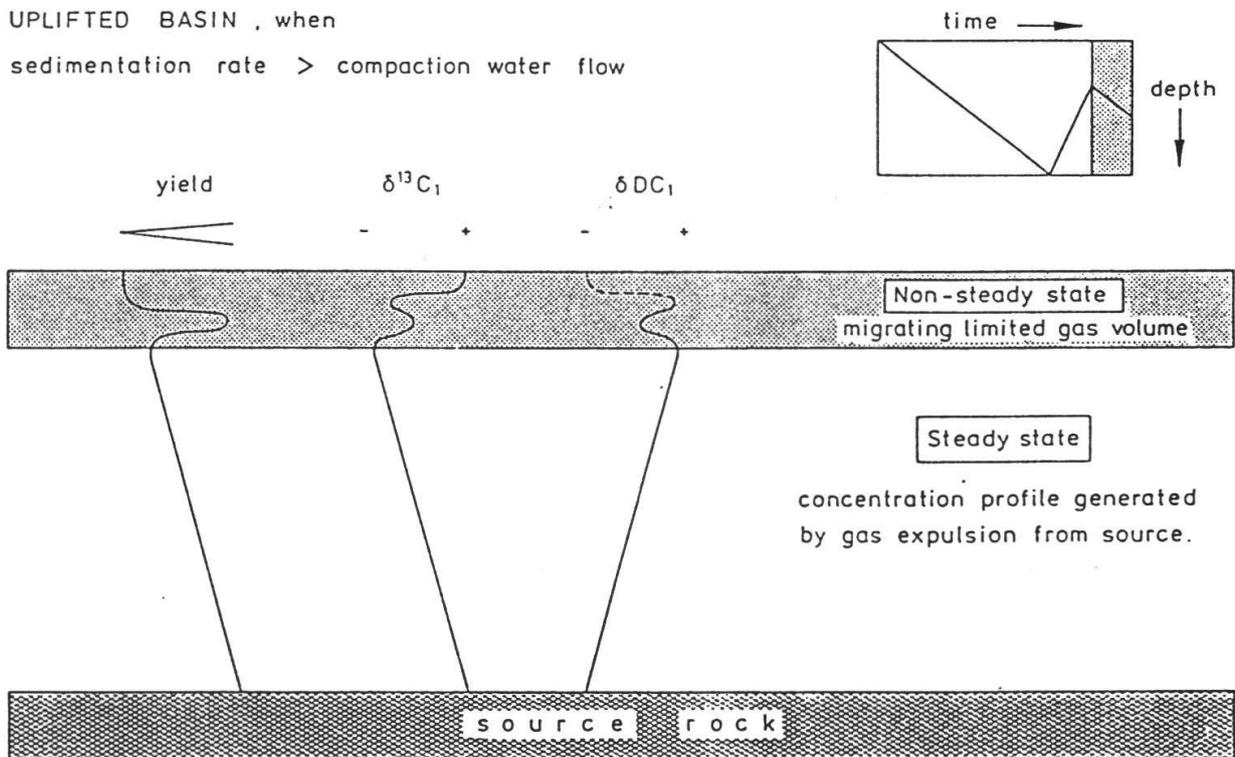


Fig. 7 Gas parameters model for uplifted basins

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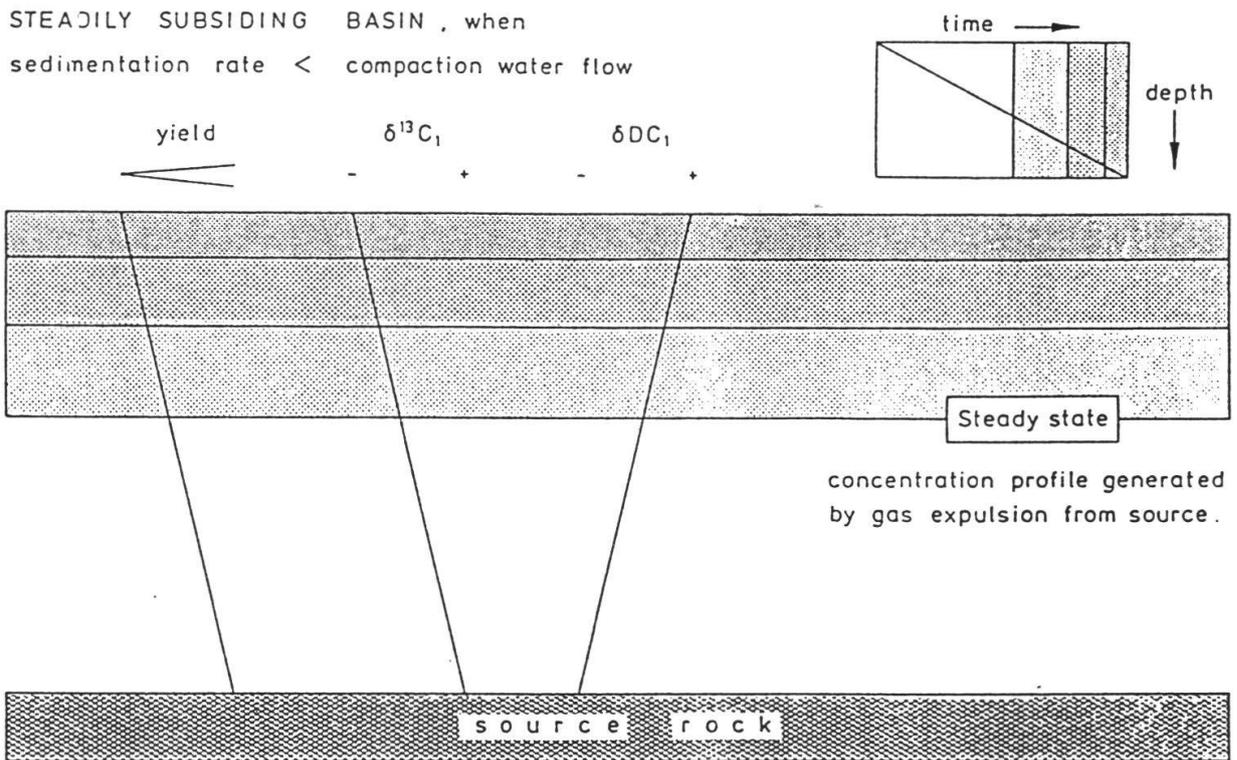


Fig. 8 Gas parameters model for steadily subsiding basins

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When the compaction water flow is greater than the sedimentation rate — which is normal in steadily subsiding basins — a spontaneous signature transmission (not more than  $10 \times 10^3$  a for basins of average depths) to the surface is guaranteed (COLEMAN et al., 1977, KETTEL et al., in preparation). This reveals in a steady-state gas concentration profile throughout the sedimentary cover, and results in a good reliability of the surface data to the subsurface signatures (Fig. 8).

It occurs sometimes in uplifted basins, specially when covered by glacial drift sediments, that the compaction water flow is smaller than the sedimentation rate. In these cases, the nearsurface layers can show a non-steady state migrating gas front, which results in significant variations of gas parameters with the sampling depth (Fig. 7). This can become more severe, when young gas migration does not fall in an era of basinal subsidence or active hydrocarbon generation.

In these cases, sampling of older layers — where outcropping — is recommended, in order to meet steady-state conditions.

On the other hand, uplifted basins — where recently eroded — could offer the unique opportunity to detect hydrocarbon accumulations directly by mapping factor A or by mapping the concentrations of heavier gases (SANDY, in press). This is because in this geological setting mostly no young hydrocarbon supply or replenishment to the accumulations from the source rocks occurs, so that the product of hydrocarbon supply x geological time for surface sediments is much higher over an accumulation than away from the accumulation. This can result in optimal variations of factor A or concentrations of heavier gases respectively.

A = direct proportional to :

- $\Delta G$  differential gas generation ( $\text{cm}^3/\text{g TOC} \times 0,1 \text{ VR}$ )
- TOC total organic carbon (%)
- $\gamma$  source rock density ( $\text{g}/\text{cm}^3$ )
- $h_1$  source rock thickness (m)
- $\Delta \text{VR}$  maturity increment in time t (%)
- q adsorption probability at temperature + 10° C
- $\mu$  dynamic viscosity of water of temperature T (cp)

F a c t o r A

A = invers proportional to :

- K permeability of sample (darcy)
- $\phi$  effective porosity of sample (%)
- t time available for gas migration (a)
- $h_2$  gas migration distance (m)
- T sample temperature (°C)
- $\Delta p/L$  pressure gradient (atm/m)

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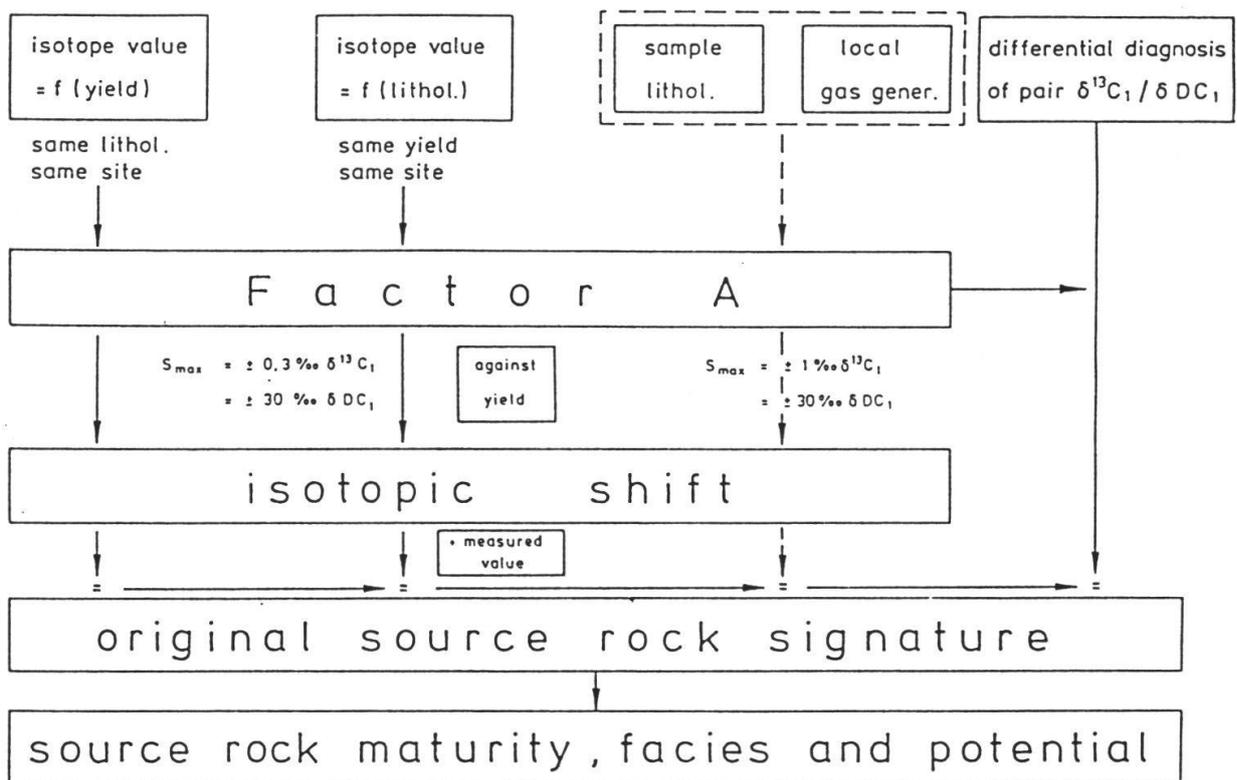
**Fig. 9** Parameters influencing Factor A

$$\text{HC supply} = \text{const.} \times A$$

( lithology, temperature, pressure gradient, migration time = const. )

Fig. 10 Possibility of the detection of hydrocarbon potential

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Fig. 11 Interpretation procedure



