

## Discussion on aluminium loss during sandstone diagenesis

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**Norman Oxtoby & Jon Gluyas** write: Wilkinson & Haszeldine (1996) have added to the lively debate on the mobility of elements during the diagenesis of silicate minerals in sandstones. They concluded from a petrographic study of the Fulmar Formation (Jurassic, North Sea) that large quantities of aluminium (Al) have been lost from the sandstone during the course of its burial history. While we have no objection to the notion that large volumes of a variety of elements can be moved in or out of sandstones (cf. Evans 1990; Gluyas & Coleman 1992\*), we believe that the evidence presented by Wilkinson & Haszeldine is ambiguous. We develop equally plausible interpretations of their data which lead to conclusions of aluminium content stasis or even import!

According to our reading of their paper, Wilkinson & Haszeldine (1996) present the argument that, *if* the initial composition of the Fulmar Formation was >35% feldspar, *then* a mass balance of Al-bearing minerals quantified from petrographic studies shows that  $c. 1500 \mu\text{mol cm}^{-3}$  Al has been exported from the most deeply buried sandstones in the studied area. We show below that we need to be more certain about the validity of the 'if' condition, before accepting many of the conclusions of this paper, including major Al loss. We do this by commenting on the statements made in the conclusions, shown in italics in the following.

(1) *The Upper Jurassic Fulmar Formation sandstones show a steady decline of K-feldspar abundance with increased burial.* This is partially correct—they show a decline from 3.2 to 4.5 km, below that the percentage appears to remain roughly constant. However, the statement uses the phrase 'with increased burial', which implies a causal relationship between burial and the dissolution of K-feldspar. There are however alternative interpretations of this correlation. Our preference, which we reveal in the course of this discussion, demands a radically different cause for the variation in K-feldspar abundance.

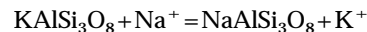
(2) *This is not matched by the precipitation of Al-bearing authigenic minerals.* This is partially correct—it requires the qualification—assuming that initial K-feldspar percentages were above 35%.

(3) *Sedimentological and provenance studies (Howell et al., 1996) suggest that the sands were of uniform composition at the time of deposition, with 35% or more feldspar.* We take issue with the use of this information. Howell et al. (1996) do not tell us anything about the feldspar percentage. In fact Howell et al. (1996) present a sedimentological model for the Fulmar Formation based on a description of sedimentary structures, trace fossil associations, sand-matrix ratios and grain size variation from core. There is no detailed mineralogical information. They conclude 'sediment was introduced into the basin by small, poorly developed fluvial systems draining the Western

Platform and Forties–Montrose high'; was deposited between steep, fault-controlled coastlines; and was redistributed by storms. Wilkinson & Haszeldine (1996) say the Fulmar sand has a point source (p. 657); Howell et al. (1996) imply a number of sources. Howell et al. (1996) also say 'Complex structure, poorly understood sedimentology and poor biostratigraphic control make the Upper Jurassic Humber Group of the South Central Graben one of the least understood and most complex hydrocarbon reservoirs of the North Sea'. They recognize 15 facies, of which six have sand contents above 80%. Wilkinson & Haszeldine (1996) claim that this shows that the sands were of uniform composition. We disagree. Clelland et al. (1993) give information on modal compositions for the Ribble Formation in the Fulmar field. This is a sandstone unit lying above the Heather and Fulmar Formations, which Clelland et al. (1993) say is mineralogically and diagenetically similar to the Fulmar Formation. This paper shows that detrital K-feldspar percentages vary between 4.7% and 14.7% (their table 1) in a 230 ft interval in a single well, with some sandstones being poorly cemented and some being well cemented. Their fig. 5 shows that the sandstone compositions vary between subarkoses and lithic arkoses. This is not uniform composition.

Wilkinson & Haszeldine (1996) have focused on the depth relations, but inspection of the geographical distribution of data points reveals that Quad 30 contains the high feldspar (quartz arenites tending to subarkoses) and Quad 29 the low feldspar (arkoses tending to subarkoses) occurrences, and that these occurrences are at least 50 km apart. Could this not be an indication of geographical variation in initial composition or intensity of near-surface processes, despite the appeal to Howell et al. (1996)?

However, let us suppose that the differences are actually due to K-feldspar dissolution. Does this imply Al-export? An alternative approach to calculating initial K-feldspar is to use the chemical mass balances from equations for albitization and illitization. Albitization is presumed to be relatively straightforward:



The illitization reaction has been formulated in a number of ways; we adopt the reaction presented by Giles & De Boer (1990) for the sake of argument:



In albitization, 1 mole of K-feldspar produces 1 mole of albite and in illitization 5 moles of feldspar are required to produce 1 mole of illite. Therefore, according to the Giles & De Boer (1990) formulation, and substituting molecular masses and densities, we need about one volume of K-feldspar to produce one volume of albite, and about two volumes of K-feldspar to produce one volume of illite. Following this calculation through, and using both albitization and illitization reactions to recalculate the initial volumes of K-feldspar, we

\*An incidental note: Gluyas & Coleman (1992) never said that shales were the source of solutes (silica)—they said that although they represented a ready source of silica, there was no positive evidence for, and a good deal of evidence against—and that researchers should do some more work on diagenetic processes.

find that the Fulmar samples initially contained between 26 and 48% K-feldspar. As an aside, this formalism also predicts that the Fulmar sands will contain between 1 and 10% diagenetic quartz. However, the main point we are making is that we can generate plausible scenarios where Al is conserved. Our argument is thus: *if* the initial composition of the Fulmar Formation was 25–50% feldspar, *then* a mass balance of Al-bearing minerals quantified from petrographic studies shows Al can be conserved. Clearly a lot of K-feldspar is destroyed in both our process and Wilkinson & Haszeldine's. Actually, it is possible to envisage scenarios where less initial K-feldspar is required, several of which involve Al-import!

Therefore, based on the data presented, it cannot be safely concluded that wholesale Al-export out of the sandstone has occurred.

(4) *Where these sandstones are buried in excess of 4.5 km, they preserve only 5% of feldspar, with a maximum of 10% illite and authigenic albite.* According to their table 1, Wilkinson & Haszeldine have 6% K-feldspar accompanied by 10% illite and 6.9% albite in 29/5b-6 at over 5.3 km, which is not what they conclude, but this is a relatively minor criticism.

(5) *Up to 1460  $\mu\text{mol}$  has been exported.* Granting their assumptions (which we do not), this follows.

(6) *The majority of the porosity within the Fulmar Formation is secondary in origin, from feldspar dissolution.* Wilkinson & Haszeldine state 'the volume of feldspar that has dissolved . . . is an order of magnitude higher than that which would have been obtained using preserved secondary porosity as an estimate of past feldspar content'. Therefore, given that they estimate about 30% loss, then preserved secondary porosity is about 3%. However, they also indicate that the Fulmar sandstones act as economic hydrocarbon reservoirs. Usually, economic oil reservoirs require at least 5% porosity, while economic oil reservoirs containing illite, which severely degrades permeability, require substantially more. From this we conclude that most of the Al-bearing diagenetic minerals, which can reach up to 30%, occlude primary porosity. We would like some clarification on how this might square with the statement in italics.

(7) *Petrographic assessment of secondary porosity underestimates feldspar dissolution by an order of magnitude.* Possibly, but given the uncertainties discussed above, not certainly.

(8) *The apparent import of quartz into these and other oilfield sandstones can be re-interpreted as quartz remaining after extensive feldspar dissolution and export of Al and other elements.* As indicated above, quartz can be generated through feldspar dissolution without Al export. Again, this may possibly be the case for the Fulmar Formation, but has yet to be demonstrated by similar studies for other sandstones. At the very least other sandstones will have to have similar initial feldspar contents.

(9) *The bulk composition of deeply buried sandstones can change significantly: from arkoses to quartzites.* In general, this seems a reasonable statement, and many studies have suggested that this has happened. *Reconstructions of sandstone or psammite provenance are likely to be in error unless they recognise this possibility.* It can go the other way. If we accept these conclusions, then we might be tempted to include some K-feldspar when there was in fact none, and end up equally in error.

In their final sentence, Wilkinson & Haszeldine plump for organic acids as the agent promoting massive dissolution of feldspar in a system with limited water. They dismiss the oft-quoted alternatives of meteoric water flushing and/or

convection cells without providing any evidence one way or another. We have no preference for cause, but would expect Wilkinson & Haszeldine to comment on points such as:

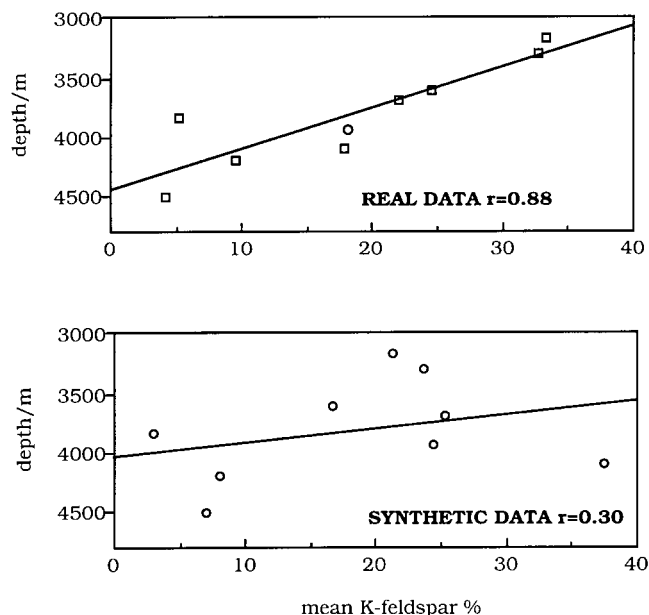
- is there a relationship between the degree of feldspar leaching and the presence or absence of petroleum in the sandstones?
- what is the dissolved aluminium content of the present formation waters?
- if the aluminium content is, as we suspect, low, where has it all gone?
- what species of organic acids or salts are dissolved in the formation waters and what are their concentrations?
- do oxygen and deuterium isotope data for formation waters confirm or rule out the presence of meteoric water in the pore system?

To conclude, we feel that Wilkinson & Haszeldine (1996) have raised some interesting ideas, some of which are plausible and which might happen under some circumstances, and may even be responsible for the effects they describe for the Fulmar Formation in Quads 29 and 30. However, we suggest that the data and arguments presented do not demonstrate this because the fundamental assumptions are not sufficiently constrained. Consequently, aluminium may not have been lost from the sandstone at all.

29 February 1997

**Mark Wilkinson & R. Stuart Haszeldine** reply: Oxtoby & Gluyas make the usual criticism of studies which compare sediments at different burial depths in order to elucidate the diagenetic processes therein. Obviously, a comparison of diagenesis at different locations is only valid if the sediments were initially of uniform composition. It is correctly stated that, in the case of the Central Graben, there is a geographic separation between the Fulmar Formation in deeply buried (Quad 29) and the more shallowly buried wells (Quad 30). However, this is normal in sedimentary basins, where deeply and more shallowly buried wells are not usually spatially admixed.

It is, however, possible to test whether the correlation between K-feldspar abundance and burial depth (3000–4500 m) is likely to be significant (Fig. 1, upper). According to our theory, in which the feldspar dissolution reaction is causally related to burial depth, so the correlation between depth and dissolution represents the reaction progress by depth and a strong relationship between depth and K-feldspar abundance would be expected (Fig. 1, upper). However, if the view of Oxtoby & Gluyas is correct, then the K-feldspar-depth plot should show nothing more than two clusters of data, one for Quad 29 and one for Quad 30. So how significant is the correlation between K-feldspar abundance and depth for the crucial depth interval at which K-feldspar dissolution is taking place? The correlation coefficient for the nine measured data points between 3200 and 4500 m is 0.88 (Fig. 1, upper). The probability of the depth–K-feldspar correlation line arising by chance is hence less than 1% ( $p$  value less than 0.01). Note that, of the nine data points on Fig. 1 (upper), 8 are from Wilkinson & Haszeldine (1996) and 1 is from well 30/16-FA31 reported in Clelland *et al.* (1993). See below for an additional discussion of the latter data. Figure 1 (lower) also illustrates a typical synthetic data set consisting of two groups of random data, one of which has the same mean and variance as the real data from Quad 29, the other the same mean and variance as the real data from Quad 30. It is clear that this does not have a



**Fig. 1.** Top: measured K-feldspar abundance (point-counted means of 3–15 analyses) versus depth. Data are from Clelland *et al.* (1993, circle) and Wilkinson & Haszeldine (1996, squares). Lower: a synthetic data set for the depth interval 3000–4500 m. This data set consists of two groups of random data, one of which has the same mean and variance as the real data from Quad 29, the other the same mean and variance as the real data from Quad 30. It is clear that the mean abundance of K-feldspar in the real data set correlates well with depth. The probability of such a correlation arising by chance is less than 1%. The synthetic data set illustrates a typical 'correlation' arising by chance, which with a correlation coefficient of only 0.30 is not significant at any reasonable confidence level.

statistically significant K-feldspar abundance–depth correlation ( $r=0.36$ ). While it would clearly be desirable to have a larger data set, we feel that this justifies our assertion that the feldspar decline is causally related to depth, and is not simply an artefact due to two spatially separate geographical areas having different sedimentary provenances.

It is interesting to note that the outlier from the K-feldspar–depth correlation (Fig. 1, upper, with 5% feldspar at 3900 m depth) is geographically removed from the remainder of the data (well 29/2a-2; fig. 1 in Wilkinson & Haszeldine, 1996). Without this point, the correlation coefficient for the K-feldspar–depth trend becomes 0.98. This well is the only one of the study wells within Quad 29 to contain kaolinite, suggesting a different early diagenesis to the remainder of the study wells. Lying as it does close to the western shelf of the graben, it has probably been subjected to meteoric water flushing during early and mid diagenesis, as have several other wells on the Western Terrace (Fig. 1, Wilkinson & Haszeldine 1996, and unpublished data). Earlier work (Wilkinson *et al.* 1994) has shown that the evidence for meteoric water flushing within the more basinwards wells of the graben in Quad 29 is at best equivocal.

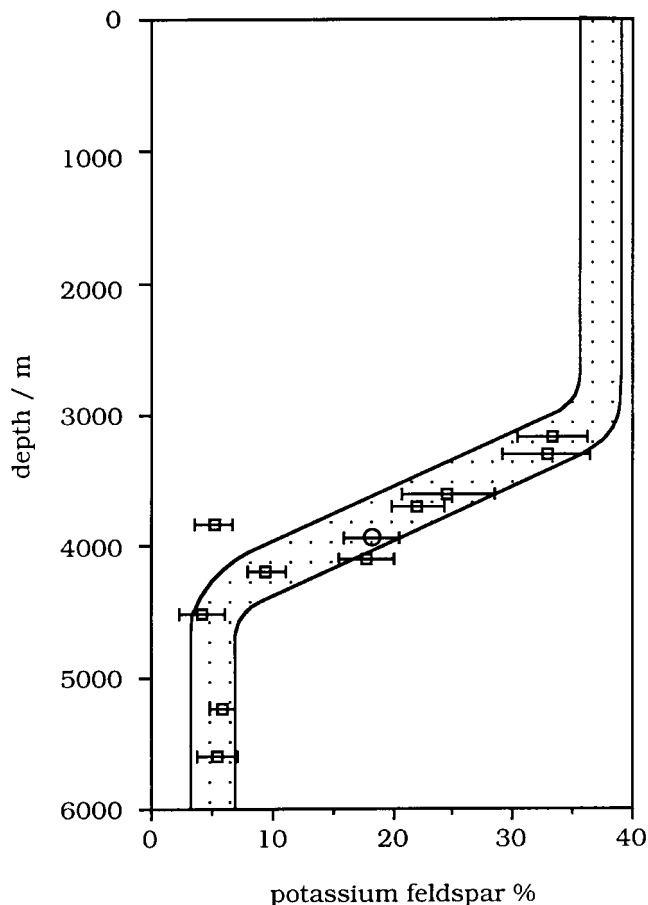
We are not claiming that every sand in the Central Graben was of identical initial composition, merely that the Fulmar Formation was sufficiently uniform over our study area to enable a meaningful comparison to be made. In fact, we know that the Fulmar Formation to the north and east of the Josephine high (Quad 23, north of Quad 30, fig. 1 of Wilkinson

& Haszeldine 1996) is of slightly different average composition. This is unsurprising as this sub-basin is separated from our study area by the Forties–Montrose–Josephine High axis.

There seems to be some confusion within the discussion of Oxtoby & Gluyas as to the importance of a sandstone *average* composition, versus that of the *individual* samples. The data on our fig. 4 (Wilkinson & Haszeldine 1996) show average sandstone compositions, expressed as a percentage of solids. The  $2\sigma$  error bars are also shown, allowing the reader to deduce for themselves how reliable the data are likely to be. For the purposes of Al mass-balancing, it is only necessary that the *average* composition of the sands were uniform at the time of deposition. Oxtoby & Gluyas cite the point-count data of Clelland *et al.* (1993), which do indeed show that the composition of individual samples within the Fulmar sands are variable on a small scale. This is not especially surprising, as published data on sandstones invariably show this degree of variation amongst individual samples (e.g. Giles *et al.* 1992, fig. 27). However, we are not concerned with the composition of individual thin sections, but with the *average* compositions of the Fulmar Formation sandbodies at particular burial depths. Taking the above example, the data of Giles *et al.* (1992, fig. 27) for the individual samples of the Brent Group shows a similar range of feldspar contents to *individual* samples to the Fulmar Formation. Even so, the decline in average feldspar content with depth is undeniable. Oxtoby & Gluyas suggest that the fact that Howell *et al.* (1996) recognize six sandstone facies within the Fulmar Formation is a problem. Yet Giles *et al.* (1992) recognize 10 different *sandstone* facies and still the feldspar decline with depth is clearly apparent!

Oxtoby & Gluyas suggest that the data of Clelland *et al.* (1993) is not compatible with our hypothesis and may amount to a disproof. Consequently, as a reply, we have included the data of Clelland *et al.* (1993) from well 30/16-FA10 in Figs 1 and 2, where it is distinguished by the plotting symbol. This additional data point slightly improved the K-feldspar–depth correlation over the interval 3000–4500 m. For some reason, Oxtoby & Gluyas cite the range of K-feldspar abundance in well 30/16-FA31 as 4.7 to 14.7%. In fact, there is a further category of point-count data labelled as 'altered feldspar', which since plagioclase is clearly rare, is most probably K-feldspar as well. This is supported by our own SEM studies. There is also a 'microcline' category, which is also K-feldspar. Summing these categories increases the abundance of K-feldspar to 9.0–20.7%. This is expressed as a percentage of whole-rock volume, and must be altered to a percentage of solids if it is to be compared to our own data. This yields a range of 11.2–28.6% K-feldspar, with a mean of  $18.4 \pm 2.3\%$  ( $2\sigma$ ). Note that this is a maximum figure, as if any of the 'altered feldspar' is in fact not K-feldspar then this mean will be decreased slightly. Reference to Fig. 1 will reveal that a slight decrease in the mean of the 30/16-FA10 K-feldspar abundance data will, if anything, improve the K-feldspar–depth correlation. Thus the data of Clelland *et al.* (1993) do not refute our hypothesis, but in fact strongly support it.

Oxtoby & Gluyas take issue with the wording of our first conclusion, that K-feldspar shows a steady decline with burial depth. We stand by our conclusion over the burial depth range of 3200–4500 m. This depth range is clearly stated both in the caption to Fig. 2 and the text of the original paper. Clearly, the sandstones could not go on losing feldspar at the same rate indefinitely, as there would soon be nothing to lose! Similarly, above 3000 m the K-feldspar abundance cannot increase to much above 35–40%. Instead, we can anticipate the shape of



**Fig. 2.** The K-feldspar–depth plot for the Fulmar Formation extended to the surface to illustrate the expected sigmoidal trend. This is as predicted for a kinetically controlled mineral transformation. The data are from Clelland *et al.* (1993, circle) and Wilkinson & Haszeldine (1996, squares). Error bars are  $2\sigma$ .

the feldspar–depth curve as being actually sigmoidal (Fig. 2), in the same manner as the smectite-to-illite transition plot of Hower *et al.* (1976) from the Texas Gulf Coast as modelled by Wilkinson *et al.* (1992, fig. 6). This is the expected shape for a reaction which is kinetically controlled. In other words, the feldspar-to-illite reaction (or, analogously, the smectite-to-illite one) proceeds only slowly at shallow depth due to low temperatures. As the sediment is buried, so temperatures and hence reaction rates increase exponentially. However, although the reaction rate continues to increase (in terms of rate per unit surface area of mineral), the reacting mineral eventually is consumed leading to a decrease in the overall rate of mineral transformation. As this is what is observed within the Fulmar Formation sandstones, the shape of the K-feldspar–depth plot is quite consistent with the large scale loss of feldspar from the sandstone.

Oxtoby & Gluyas attempt an alternative calculation of aluminium mass balance, and claim that two volumes of K-feldspar produce one volume of illite. This leads them to propose that Al is conserved within the sandstone. However, Oxtoby & Gluyas have made a serious error of omission in their Al mass balance calculation: they have not allowed for micro-porosity within the illite. As is well known, authigenic illite does not form dense masses of interlocking crystals, but forms fibrous or lath-like crystals ('hairs') which surround

**Table 1.** Mineral parameters for Al-conservation calculation

Mineral	Molar mass (g)	Density ( $\text{g}/\text{cm}^3$ )*	Microporosity (%)
K-feldspar	278.3	2.55–2.63	0
Illite	722.6	2.6–2.9	50†

\*From Deer *et al.* (1982).

†Our own conservative estimate.

large volumes of microporosity. When a rock sample is point-counted, this microporosity is counted as illite, especially if (as is the case with the Fulmar Formation) the microporosity is impregnated with bitumen. We use a conservative estimate of the microporosity content of illite as 50%.

Hence, when Oxtoby & Gluyas calculate that two volumes of K-feldspar will produce one volume of (point-counted) illite, they are wrong. In fact, two volumes of K-feldspar will produce *two* volumes of (point-counted) illite, as 50% of this 'illite' is in fact porosity. Although this calculation is simple, we reproduce it here to prevent further confusion. We use the feldspar decomposition reaction of Giles & De Boer (1990) which was reproduced by Oxtoby and Gluyas, such that 5 moles of K-feldspar will produce a single mole of illite. The molar masses, densities and microporosities of these minerals are given in Table 1. The molar volume of a mineral is given by:

$$\text{Molar volume} = \text{molar mass}/\text{density}$$

This gives 106–109  $\text{cm}^3$  for K-feldspar, 249–278  $\text{cm}^3$  for *solid* illite, the uncertainty arising from the variation within the published figure for density. Hence 5 moles of K-feldspar (530–545  $\text{cm}^3$ ) will generate 1 mole of *solid* illite (249–278  $\text{cm}^3$ ). We must now add in the microporosity, using the conservative estimate of 50% from Table 1. This means that 5 moles of K-feldspar (530–545  $\text{cm}^3$ ) will generate 1 mole of *point-counted* illite (498–556  $\text{cm}^3$ ). It follows that 1 volume of K-feldspar will generate approximately 1 volume of illite as it is point-counted, and not only a half volume as stated by Oxtoby & Gluyas. The correct ratio of K-feldspar to illite has been incorporated into the 'closed system trend' of our fig. 3 (Wilkinson & Haszeldine 1996).

There is one further point to consider. When point-counting 'illite', it is very difficult to distinguish between detrital and authigenic pore-lining clays. In fact, in the fine-grained, bitumen-impregnated Fulmar Formation, it is impossible. The point-counted 'illite' figures we have used in our calculation will hence include any detrital clay present. The 'illite' percentages are hence *maxima*, and the real illite contents of the sandstones will be lower. Examining our original fig. 3 (Wilkinson & Haszeldine 1996) this will tend to move all the plotted points to the left, i.e. away from the line of Al conservation! We therefore stand by our original conclusion, that (assuming an originally uniform *average* composition) the Fulmar Formation has exported Al during burial from 3 to 4.5 km depth.

Concerning the origin of the porosity within the Fulmar Formation, Oxtoby & Gluyas take issue with our assertion that the majority of the porosity within the deeper wells is of secondary origin and that this is not apparent from point-count analysis. We consider that both primary and secondary

porosity are destroyed by compaction and cementation during burial. As compaction is most effective at shallow burial depths, and primary porosity is not renewed, then secondary porosity (which is forming most rapidly between 3200–4500 m) has the best chance of survival. That the majority of the cements may be infilling primary porosity, as suggested by Oxtoby & Gluyas is not a problem for us—we were not talking about minus-cement porosity, but about open porosity. We re-iterate our point that, as point-counted secondary porosity levels in the Fulmar Formation sandstones are only between 0.5 and 4.5% average (M.W. and R.S.H. unpublished data), they are not a reliable estimate of the volume of feldspar which has undergone dissolution. We suggest that the distinctive and discriminating petrographic characteristics of secondary porosity are not preserved within these newly-formed pores. Any estimate of missing feldspar based on the petrographic recognition and point-counting of secondary porosity could be an order of magnitude out! The other conclusion with which Oxtoby & Gluyas take issue is the relevance of large-scale feldspar dissolution to provenance reconstruction. This still seems obvious to us.

We admit that our data do not prove *by what mechanism* Al was exported from the Fulmar Formation. This was never the purpose of the paper. Instead, the aim was to show that it is probable that Al export has occurred, and by inference, that an export mechanism must exist. Oxtoby & Gluyas misrepresent our wording—all we stated was that organic acids remain the only known mechanism for enhancing the solubility of Al in the sub-surface. We do not have the analytical data on porewaters that Oxtoby and Gluyas request, and it is not our intention to be side-tracked into a (dataless) argument about the viability of organic acids as Al complexing agents. This clearly remains an interesting problem for future research.

We thank Oxtoby & Gluyas for giving us this opportunity to further explain our views, and for drawing our attention to the data of Clelland *et al.* (1993). We consider that we have robustly shown and re-affirmed that it is highly probable that

the Fulmar Formation has exported Al during burial, and that the quantity of secondary porosity within the sandstones is significantly greater than would be surmised by simply using point-count data.

24 February 1997.

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