

Letter to referee 1

The paper has been modified according to the specific requests of the referee.

In particular, Sections 4 and 5 –which were found to have the most significant problems- have been substantially modified and some parts have been totally rewritten.

The paragraph '4.2 Flow characteristics' has been rewritten : a part has been added about the linear and the non linear terms A and B and the correspondent flow rate at which inertial terms are dominant has been determined and compared with other people's results. Some considerations have been added about when inertial terms cannot be negligible in the reality.

In paragraph '4.3 Solute transport' a new figure (Fig 10) has been added to enhance the concept that there is a pronounced mobile immobile zone interaction cannot be negligible and that the presence of a transitional regime affects the velocity field.

'As the flow rate increases the difference between transport time and exchange time decreases and for high values of flow rates they get closer to each other. Furthermore in analogy with Figure 9 when inertial forces begin to become dominant ($FO \geq 1$) a change in the slope of the relationships a) transport time vs. flow rate and b) exchange time vs. flow rate can be evidenced as a consequence the non – equilibrium behavior becomes stronger.'

The increase in mass exchange coefficient with velocity has been interpreted as due to higher mixing in the mobile phase at high pore-water velocities or as due to shorter diffusion path lengths as a result of a decrease in the amount of immobile water. The results obtained have been compared with other results in the literature (van Genuchten and Wierenga, 1977; Nkedi-Kizza et al., 1983; De Smedt and Wierenga, 1984; De Smedt et al., 1986; Schulin et al., 1987).

The absence of effects of inertia on dispersion and the prevalence of geometric dispersion have been explained, have been commented in terms of pollutants' propagation and have been compared with other people's results:

'These experimental results suggest that geometrical dispersion dominates the effects of Aris – Taylor dispersion. This can be attributable to two possible mixing processes: the presence of a transient regime that precedes stationary dispersion and the presence of complex paths that enhance the geometrical dispersion and do not allow the development of Aris –Taylor dispersion.

In order to get the Aris - Taylor dispersion regime completely established the solute must travel over a minimum distance much larger than the product of velocity and the characteristic time of transverse diffusion (Equation 6). In our case considering a value of molecular diffusion equal to $D_m \approx 1.85 \times 10^{-9} \text{ m}^2\text{s}^{-1}$ the ratio between the minimum distance and the length of active path is in the range 2.88 – 19.61, therefore according to Berkowitz and Zou (1996) the Aris – Taylor dispersion coefficient is time dependent.

In the case study, due to the complex tortuous topology of the fracture network there may not be an opportunity for Aris – Taylor dispersion to develop. These results need to be compared with previous studies carried out in limestone, granite and welded tuff fractures (Kumar et al, 1995, Yeo et al, 1998, Wan et al, 2000) that found the dominance of Taylor dispersion. In most fractures used in previously

reported experiments (Detwiler 2000), the fraction of contact area between the fracture surfaces was relatively small. The resulting long, simply connected flow paths permitted Taylor dispersion to “develop”. In the case study, due to the complex tortuous topology of the fracture network there may not be an opportunity for Taylor dispersion to develop.'

The conclusions have been totally rewritten and the results have been better interpreted in hydrogeological terms (the MIM better explains BTC curves in fractured media because it is physically based, the presence of inertial effects has been interpreted as mass transfer limitation, recirculation effects and therefore delay in mass transport; the effects of this delay has been interpreted in terms of aquifer remediation.

'The presence of not negligible inertial effects plays an interesting role on the structure of the flow field and can lead to the presence of recirculation zones that enhance the non – equilibrium behavior. These recirculation zones represent low velocity or stagnant region that represents the physical immobile zones. The fact that in the present study the Damköhler number is close to the unity especially for high flow rate and the exchange coefficient depends on the flow velocity could confirm this hypothesis.'

'Mass-transfer limitation can have a significant impact on the mobility of contaminants in groundwater (Maraqa, 2001) and therefore on the predicted cleanup times. Berglund & Cvetkovic (1995) proposed an analytical solution for the radial mass flux in a heterogeneous aquifer, which refers to a simple case of aquifer remediation by the pump and treat method using an extraction well. The results showed a large initial drop in concentration followed by a leveling and a very slow gradual decline. The general effects of the rate - limited mass transfer process are a considerable increase in cleanup times and that cleanup times tend to converge towards constant values, where further increase in pumping rate has no effect in terms of reduced cleanup times. They concluded that the advantages of using high pumping rates are small if the recovery of the contaminants in the immobile phase depends on a mass transfer process characterized by a large time scale.'

In the conclusion the study is aimed at interpreting the obtained results in hydrogeological terms, as requested by the referee.

'Does the result say anything about contributions to immobility of the fracture stagnation zones or the internal pores of the limestone, or does it have significance for pollution or remediation issues, or does it provide guidance on how such studies should be conducted in the future? The paper needs to convince readers that what has been learned here is new and important.'

'In fractured aquifers, the prolonged BTCs can also be attributed to intermediate storage in zones containing quasi-immobile groundwater, and slow release into active fractures. These phenomena demonstrate that fractured aquifers are not always fast-flushing systems, but contaminants can sometimes remain in immobile fluid regions for long periods (Goldscheider, 2008). The persistence of some pollutants creates a costly, remedial challenge in the subsurface.'

Due to the complexity of hydrogeologic conditions in tight formations, setting up a decontamination treatment in fractured rock aquifers is a complex undertaking: traditional remediation processes adopted for porous media clean up do not work well for the removal of most pollutants. Groundwater extraction and treatment systems are not fully effective in speeding up the aquifer's remediation,

because of diffusive processes that tend to retard a contamination plume's advance through a fractured rock aquifer and to substantially increase the difficulty of purging pollutants from it.

Therefore, the traditional remediation techniques have to be combined with new innovative techniques that prove to be effective in speeding up the aquifer's cleanup.

In classic depollution strategies such as Pump & Treat system, an induced-hydraulic gradient much higher than natural gradient is often necessary to clean up contaminated groundwater, where non-Darcian flow is expected to occur. Under these circumstances, the behavior of solute dispersion needs to be evaluated thoroughly for the full range of flow regimes.'

This study motivates further experimental work to increase our understanding of the controlling mechanisms and key parameters for flow and transport in fractured media, and thus make better predictions of the infiltration path of a solute, the location of trapped contaminants, the effect of containment or mobilization techniques for pollutants, and the remediation of a contaminated fractured aquifer.'

The English has been improved and some expression as 'in correspondence of' has been substituted with 'at' or 'beside' or 'along'.

The list of references has been completed and some more references have been added.

The variable h_c has been defined and the equation numbers have been corrected.

In figure 3 h_c , a and b have been inserted.

'Difference head' has been corrected with 'hydraulic head difference'.

Figure 9 has been better explained in the caption.