



Comparative evaluation of ozonation and stripping methods to treat contaminated groundwater by trichloroethylene. Assessment of effects on the other matrix components



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ABSTRACT

Many methods, that involve several oxidants combined specific catalyst, are commonly utilised for the abatement of organochlorine compounds in natural and waste waters. This study investigates the efficiency of removal of trichloroethylene (TCE) contained in groundwater through oxidation with ozone. The fundamental contribution of the stripping was experimentally measured through tests with nitrogen. For comparative purposes, the same experimental tests were performed on a model solution containing TCE. A dedicated experimental apparatus was specifically designed. The treatment under investigation showed a good efficiency dependent on the ozone dose and initial pollutant concentration. Experimental data, obtained from stripping and oxidation tests performed both with groundwater and model solution, were fitted by models. Finally, the effects of the ozone treatment on other groundwater components, as bromide and organic and inorganic nitrogen, together with the formation of undesired by-products were also taken into account.

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Introduction

The environmental contamination by halogenated solvents such as trichloroethylene and the associated risks to humans represent a public health threat [1]. Due to its wide use, historical improper disposal and low tendency to degradation, TCE is a widespread polluting agent in groundwater (GW) [2] and since long time it has attracted the attention of world organizations and scientists [3]. Since 1995 the IARC (International Agency for Research on Cancer) classified TCE in group 2A (probable human carcinogen) [4] and the Europe Council Directive 98/83/EC established that the TCE concentration, on the quality of water intended for human consumption, must be lower than 10 µg/L.

The low solubility in water (1100 mg/L at saturation) and high density (1.46 g/mL) allow TCE to persist in aquifers as pools of dense non-aqueous-phase liquid, becoming sources of continuous contamination by slow dissolution into groundwater [2].

As a consequence, contamination by this halogenated aliphatic compound prevents the use of groundwater for drinking, irrigation or nearly any other beneficial use.

This type of pollution represents a serious problem in many industrialised areas of Italy. In Campania, a southern Italian region, the

widespread presence of halogenated aliphatic compounds in groundwater is an important environmental pollution problem [5].

Many methods have been proposed for the remediation of groundwater, both *in situ* and *ex situ* [5, 6, 7, 8, 9, 10]. Although these methods, such as PAB (permeable adsorptive barrier), show good result for the removal of chlorinated halogenated, their application can involve complex and expensive technical work. Among the *ex situ* treatments, the pump-and-treat (P&T) methods are most widely used. The term P&T is used in a broad sense to include any system, also mobile, where withdrawal from groundwater is part of the remediation strategy. An attractive solution against TCE pollution, as technologies for *ex situ* groundwater remediation, is the pump and oxidation by ozone. Usually, for TCE treatment the ozone has been employed in combination with other oxidant reagents and/or catalysts (for example UV, H₂O₂, TiO₂) that have the task of assisting the oxidation reaction. Although these have methods shown adequate removal, they have a high technological complexity which can lead to high costs respect to the use of the only ozone [10, 11, 12, 13].

In removal processes of compounds that involve ozone, the oxidation can occur via ozone itself or hydroxyl radicals. OH, or via a combination of both. In fact, ozone is unstable in water, where it decays through complex reaction path chains that depend on the matrix composition [14].

There is a lack of information about removal process of TCE from GW through only ozonation. Besides, the removal processes through ozonation imply a certain contribution due to the action of the stripping which is generally not properly highlighted in the experimental

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works. This study investigates the efficiency of ozone for the TCE removal from contaminated groundwater and presents an evaluation of the stripping contribution to the removal of TCE [15]. For this purpose, a range of doses of ozone and flow rates of gas were tested. For comparative purposes the investigation was also performed in purified water (PW) (reagent grade). To this aim a dedicated experimental apparatus was specifically designed and experimental tests with groundwater and purified water spiked with TCE were performed. Experimental data were fitted with models obtained by mass balance. Some parameters of models were evaluated experimentally.

Finally, because ozone and $\bullet\text{OH}$ radicals are not selective, together with the beneficial effects, undesired by-products can be formed from the oxidation of some groundwater components [16]. This groundwater samples were analysed before and after oxidation treatment, in order to verify the effects on the chemical composition of the matrix. Particular emphasis was given to the formation of bromate, nitrite and nitrate, affected by the initial TCE amounts.

Materials and methods

Experimental apparatus

Fig. 1 is illustrated the experimental apparatus. This was specifically designed as a semi-batch plant: with continuous flow for the gas and a batch for the liquid. The reactor is a plexiglas column at the bottom of which a porous diffuser plate is placed. The column has a length of 120 cm and an internal diameter equal to 5 cm. On the top of the column the link for the off-gas is placed, a liquid sampling point is located 40 cm from the bottom of the column. All valves and fittings are Teflon made. Ozone was generated by a Model Microlab 1, Aeraque S.r.l., Italy, fed with pure oxygen (99.998%). The ozone production, which can be changed in the range 1.3–7.03 mg/min, was regulated, at fixed flows of oxygen (0.1, 0.2 and 0.4 L/min), through adjustments of the generator power. Exhaust gas from the reactor was flowed through a Drechsel trap and then to a catalytic ozone destroyer. A valve system was used to switch the gaseous flow to an on-line spectrophotometer with provision for measurement at the inlet and outlet, for real-time ozone concentration monitoring. The plant was connected to a nitrogen line (99.999%) during the stripping tests.

Groundwater and purified water

The two aqueous matrices were characterised before being used in the experimental tests. A volume of 50 L of the groundwater was drawn by electric submersible pump from a 40 m deep well, in ENEA's Research Centre Portici located near Naples, Italy.

GW was filtered through a 0.45 μm Wathman membrane and analysed its chemical principal constituents by methods described in Section 2.3. The results are reported in Table 1. The PW was obtained with a Water purification System equipped with MilliQ gradient A10 plus and Elix 3 in series by Millipore S.A.S., France. The PW quality is consistent with purity standards described by ASTM[®], ISO[®] 3696 and CLSI[®] norms and has the following principal characteristics: 18 M Ω resistivity, T.O.C. < 5 $\mu\text{g/L}$, bacteria < 0.1 cfu/mL, Cl^- < 1 $\mu\text{g/L}$, and Na^+ < 1 $\mu\text{g/L}$.

Spiked solutions in PW and GW were prepared by adding the proper amounts of TCE for two concentration levels (80 and 300 $\mu\text{g/L}$).

Materials and analytical methods

Trichloroethylene 99.5% (Fluka Analytical, Switzerland) was used. According to APAT-IRSA CNR method 5150 [17], the TCE concentrations were analysed by gas-chromatography. Water samples were previously concentrated with a "purge & trap" system connected

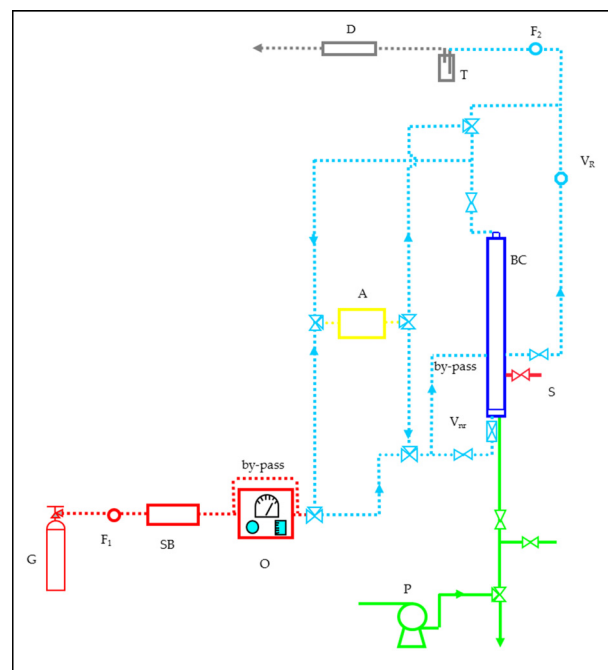


Fig. 1. Experimental equipment. A: spectrophotometer; G: gas supply; BC: bubble column; D: ozone destroyer; F1–F2: flowmeters; SB: silica bed; O: ozone generator; P: peristaltic pump; S: water sampling; T: water trap; V_{NR}: no-return valve; V_R: pressure drop regulation valve; continued line: water line; dashed line: gas line.

Table 1

Means values of main chemicals present in the groundwater used for experimental activity. The GW analysis was repeated at the beginning of each experiment.

Parameters	Concentrations (mg/L)
Fluoride ^a	3.5 ± 0.2
Bromate ^a	<0.1
Chloride ^a	109.1 ± 6.7
Nitrite ^b	0.048 ± 0.005
Bromide ^a	0.18 ± 0.02
Nitrate ^a	104.3 ± 9.2
Sulphate ^a	154.6 ± 7.6
TOC ^c	2.9 ± 0.5
Ammonia ^b	0.004 ± 0.001
Phosphate ^b	0.300 ± 0.015
pH	7.0 ± 0.2

^aBy ion chromatography.

^bBy flow injection analysis.

^cBy TOC analysis.

to a Gaschromatograph (Model 6890 N, Agilent Technologies, US) equipped with an electron capture detector (ECD). A DB264 capillary column (30 m × 0.32 mm × 1.80 μm) was used at a 2 mL/min carrier gas flux (Helium). Separation was performed with an isothermal programme at 110 °C. The detector temperature was set at 300 °C.

Anion analysis (Cl^- , F^- , SO_4^{2-} , BrO_3^- , Br^- and NO_3^-) was performed by ion chromatography with a DX 500 system by Dionex Corporation, US. An Ion Pac AS23 Dionex column was used at 1 mL/min flow rate a carbonate/bicarbonate 4.5 mM/0.8 mM solutions was used as isocratic eluent (detection limit 100 $\mu\text{g/L}$). Ammonia, phosphate and nitrite anions were determined according to APAT-IRSA colorimetric method 4030, 4010 and 4050 respectively [17], using a μMac 1000 Flow Injection Analyzer by Systea S.p.a., Italy (detection limit 1 $\mu\text{g/L}$). TOC (total organic carbon) was determined using a TOC-5000 Analyzer by Shimadzu Corporation, Japan (detection limit 0.1 mg/L). Ozone concentrations in the gas phase were analyzed using a Cary 1E spectrophotometer (Varian, Australia) at 228 nm wavelength equipped with a cylindrical flow-through quartz cell. The calibration

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