Simulation of the mantle and crustal helium isotope signature in the Mediterranean Sea using a high-resolution regional circulation model

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Abstract.

Helium isotopes (^{3}He, ^{4}He) are useful tracers for investigating the deep ocean circulation and for evaluating ocean general circulation models, because helium is a stable and conservative nuclide that does not take part in any chemical or biological process. Helium in the ocean originates from three different sources: namely, (i) gas dissolution in equilibrium with atmospheric helium, (ii) helium-3 addition by radioactive decay of tritium (called tritiugenic helium), and (iii) injection of terrigenic helium-3 and helium-4 by the submarine volcanic activity which occurs mainly at plate boundaries, and also addition of (mainly) helium-4 from the crust and sedimentary cover by α-decay of uranium and thorium contained in various minerals.

We present the first simulation of the terrigenic helium isotope distribution in the whole Mediterranean Sea, using a high-resolution model (NEMO-MED12). For this simulation we build a simple source function for terrigenic helium isotopes based on published estimates of terrestrial helium fluxes. We estimate a hydrothermal flux of 3.5 mol ^{3}He yr^{-1} and a lower limit for the crustal flux at 1.6 x 10^{-7} mol ^{4}He mol m^{-2} yr^{-1}.

In addition to providing constraints on helium isotope degassing fluxes in the Mediterranean, our simulations provide information on the ventilation of the deep Mediterranean waters which are useful for assessing NEMO-MED12 performance. This study is part of the work carried out to assess the robustness of the NEMO-MED12 model, which will be used to study the evolution the biogeochemical cycles in the Mediterranean Sea under a changing climate, and to improve our ability to predict the future evolution of the Mediterranean Sea under the increasing anthropogenic pressure.

1 Introduction

Helium isotopes are a powerful tool in Earth sciences. The ratio of ^{3}He to ^{4}He varies by more than three orders of magnitude in terrestrial samples. This results from the distinct origins of ^{3}He (essentially primordial) and ^{4}He (produced by the radioactive decay of uranium and thorium series) and their contrasting proportions in the Earth's reservoirs (Fig.1). The atmospheric ratio, $R_{air} = {}^{3}He/{}^{4}He = 1.384 \times 10^{-6}$ (Clarke et al., 1976), can be considered constant due to the long residence time of helium, which is ~ 10^{6} times longer than the mixing time of the atmosphere (based on the total helium content of the atmosphere and the global helium degassing flux estimated by Torgersen, 1989). Relative to this atmospheric ratio, typical $^{3}He/^{4}He$ ratios vary from <0.1 R_{air} in the Earth's crust to an average of 8±1 R_{air} in the upper mantle, and up to some 40 to 50 R_{air} in products of plume-related ocean islands, such as Hawaii and Iceland (Ballentine and Burnard, 2002; Graham, 2002; Hilton et al., 2000).

At the ocean surface, helium is essentially in solubility equilibrium with the atmosphere. However at depth, several important processes alter the isotopic ratio (Fig.1 - see Schlosser and Winckler (2002) for review). Firstly, ^{3}He is produced by the radioactive decay of tritium (Jenkins and Clark, 1976); and secondly terrigenic helium is introduced not only by the release of helium from submarine volcanic activity at mid-ocean ridges and volcanic centres, with elevated $^{3}He/^{4}He$ ratios typical of their mantle source (Lupton et al., 1977a, b; Jenkins et al., 1978; Lupton, 1979; Craig and Lupton, 1981; Jean-Baptiste et al., 1991a, 1992); but also by the addition of helium with a low $^{3}He/^{4}He$ ratio from the crust and sedimentary cover, mostly due to α-decay of uranium and thorium minerals (Craig and Weiss, 1971).

Oceanic $^3He/^4He$ variations are usually expressed as δ^3He, the percentage deviation from the atmospheric ratio, defined as $(R_{sample}/R_{air} - 1)100$. Below the mixed layer, oceanic $^3He/^4He$ values are usually significantly higher than the atmospheric ratio, with δ^3He up to 40% in the Pacific Ocean (Craig and Lupton, 1981; Lupton, 1998). However, there are some exceptions. Intra-continental seas such as the Black Sea and the Mediterranean display deep water $^3He/^4He$ ratios indicative of a preferential addition of ^4He-rich crustal helium rather than ^3He-rich mantle helium (Top and Clarke, 1983; Top et al., 1991; Roether et al., 1998, 2013).

Early investigations in the eastern Mediterranean (Meteor cruise M5/1987, Roether et al. (2013)) have indeed revealed that deep waters have a crustal helium signature, with δ^3He as low as -5% (Fig. 2). Note that Fig. 2 shows this deep core of crustal helium is being progressively erased by the addition of tritiugenic ^3He produced by the bomb tritium transient and by the recent dramatic changes in the thermohaline circulation of the EMed, known as the Eastern Mediterranean Transient (EMT) (Roether et al., 1996, 2007, 2014), during which dense waters of Aegean origin replaced the Adriatic source of the deep waters in the EMed.

Deconvolution of the various helium components using neon indicates that the mantle helium contribution is only ~5% (Roether et al., 1998). In the Mediterranean Sea terrigenic helium is therefore largely of crustal origin due to the presence of a continental-type crust and a high sediment load of continental origin, but also because mantle helium, which is produced by the submarine volcanic activity in only a few places in the Mediterranean Sea (Eolian Arc, Aegean Arc, Pantelleria Rift in particular), is released at rather shallow depths (Dando et al., 1999) and is therefore quickly transferred to the atmosphere.

Mantle ^{3}He was discovered in the deep ocean by Clarke et al., 1970. It is injected at mid-ocean ridges as part of the processes generating new oceanic crust, and advected by ocean currents. Since this discovery, helium isotopes have been used extensively to trace the deep ocean circulation (Jamous et al., 1992; Jean-Baptiste et al., 1991b, 1997, 2004; Lupton, 1996, 1998; Top et al., 1991; Rüth et al., 2000; Well et al., 2001; Srinivasan et al., 2004) and to study ocean dynamics (circulation, ventilation and mixing processes) in conjunction with tritium (Andrie and Merlivat, 1988; Jenkins, 1977, 1988; Schlosser et al., 1991; Roether et al., 2013). Ventilation is defined as the process of moving a parcel of water from the surface to a given subsurface location. It can occur through convection, sub-duction, advection, and diffusion (Goodman, 1997; England, 1995).

The helium isotope distribution in the deep oceans has also been simulated by various ocean circulation models to constrain global helium degassing fluxes and evaluate the degree to which models can correctly reproduce the main features of the world's ocean circulation (Farley et al., 1995; Dutay et al., 2002, 2010; Bianchi et al., 2010).

In this study we build a source function for the release of terrigenic helium components (crust and mantle) to the deep Mediterranean and apply it to a high-resolution oceanic model of the Mediterranean Sea. The simulated helium-isotope distribution is then compared with available data (see §4) to constrain terrigenic helium fluxes. In addition to providing constraints on the degassing flux, our work is the first attempt to simulate natural helium-3 in a high-resolution regional model of the Mediterranean Sea and provides new information on the model's capacity to represent the ventilation of deep waters.

2 Description of the model

The model used in this work is a free surface ocean general circulation model NEMO (Nucleus for European Modelling of the Ocean) (Madec and NEMO-Team., 2008) in a regional configuration called NEMO-MED12 (Beuvier et al., 2012a).

This model of the Mediterranean Sea has been used previously to study anthropogenic tritium and its decay product helium-3 (Ayache et al., 2015), the anthropogenic carbon uptake (Palmiéri et al., 2015), the transport through the Strait of Gibraltar (Soto-Navarro et al., 2014), as well as the Western Mediterranean Deep Water (WMDW) formation (Beuvier et al., 2012a), and the mixed layer response under high-resolution air-sea forcings (Lebeaupin Brossier et al., 2011). This model satisfactorily simulates the main structures of the thermohaline circulation of the Mediterranean Sea, with mechanisms having a realistic timescale compared to observations. In particular, tritium/helium-3 simulations (Ayache et al., 2015) have shown that the Eastern Mediterranean Transient (EMT) signal from the Aegean sub-basin is realistically simulated, with its corresponding penetration of tracers into the deep water in early 1995. The strong convection event of winter 2005 and the following years in the Gulf of Lions was satisfactorily captured as well. However, some aspects of the model still need to be improved: in the eastern basin, tritium/helium-3 simulations have highlighted the too- weak formation of Adriatic Deep Water (AdDW), followed by a weak contribution to the EMDW in the Ionian sub-basin. In the western basin, the production of WMDW is correct, but the spreading of the recently ventilated deep water to the south of the basin is too weak. The consequences of these weaknesses in the model0 s skill at simulating some important aspects of the dynamics of the deep ventilation of the Mediterranean will have to be kept in mind when analysing these helium simulations.

NEMO-MED12 covers the whole Mediterranean Sea, but also extends into the Atlantic Ocean. Horizontal resolution is one-twelfth of a degree, thus varying with latitude between 8 and 6.5 and 8 km from 30° N to 46° N, respectively, and between 5.5 and 7.5 km in longitude. Vertical resolution varies with depth, from 1 m at the surface, to 450 m at the bottom (50 levels in total). We use partial-steps to adjust the last numerical level with the bathymetry. The exchanges with the Atlantic Ocean are performed through a buffer zone, from 11°W to 7.5°, where 3-D temperature and salinity model fields are relaxed to the observed climatology (Beuvier et al., 2012a). NEMO-MED12 is forced at the surface by ARPERA (Herrmann and Somot, 2008; Herrmann et al., 2010) daily fields of the momentum, evaporation and heat fluxes over the period 1958-2013. For the sea-surface temperature (SST) a relaxation term is applied to the heat flux (Beuvier et al., 2012a). The total volume of water in the Mediterranean Sea is conserved by restoring the sea-surface height (SSH) in the Atlantic buffer zone toward the GLORYS1 reanalysis (Ferry et al., 2010).

The initial conditions (temperature, salinity) for the Mediterranean Sea are prescribed from the MedAtlas-II (MEDAR-MedAtlas-group, 2002; Rixen et al., 2005) climatology weighted by a low- pass filter with a time window of 10 years using the MedAtlas data covering the 1955-1965 period, following Beuvier et al. (2012a). For the Atlantic buffer zone, the initial state is set from the 2005 World Ocean Atlas for temperature (Locarnini et al., 2006), and salinity (Antonov et al., 2006). River runoff is prescribed from the interannual data set of Ludwig et al. (2009) and Vörösmarty et al. (1996).

Full details of the model and its parameterizations are described by Beuvier et al. (2012a, b); Palmiéri et al. (2015) and (Ayache et al., 2015).

3 The tracer model

Helium is implemented in the model as a passive conservative tracer which does not affect ocean circulation. It is transported in the Mediterranean Sea by NEMO-MED12 physical fields using an advection-diffusion equation (Eq. 1). The rate of change of the concentration of each specific passive tracer C is:

$$\frac{\delta C}{\delta t} = S(C) - U.\nabla C + \nabla.(K\nabla C) \quad (1)$$

where S(C) is the tracer source (at the seafloor) and sink (at the air-sea interface); U.∇C is advection of the tracer along the three perpendicular axes and ∇. (K∇C) is the lateral and vertical diffusion, with the same parameterization as for the hydrographic tracers.

Because ^3He, ^4He are passive tracers, simulations could be run in a computationally efficient off-line mode. This method relies on previously computed circulation fields (U, V, W) from the NEMO-MED12 dynamical model (Beuvier et al., 2012a). Physical forcing fields are read daily and interpolated to give values for each 20-min time-step. The same approach was used by Ayache et al. (2015) to model the anthropogenic tritium invasion and by Palmiéri et al. (2015) for simulating CFCs and anthropogenic carbon. This choice is justified by the fact that these tracers are passive. Their injection does not alter the dynamics of the ocean, and they have no influence on the physical properties of water, unlike hydrographic tracers such as temperature or salinity.

The simulations were initialized with uniform ^3He and ^4He concentrations corresponding to those at solubility equilibrium with the partial pressures of these isotopes in the atmosphere, for seawater at T=10°C and S=34 (Weiss, 1971). Model simulations were integrated for five hundred years until they reached a quasi-steady state, i.e., the globally averaged drift was less than 10^{-2} δ^3He % per two hundred years of run

3.1 Parameterization of the helium injection

Terrigenic helium in the Mediterranean Sea has two components: 1) Crustal helium, originating from the crust and overlying sediment cover, and 2) mantle helium, injected by submarine volcanic activity. For the injection of helium, we follow the protocol proposed by (Dutay et al., 2002, 2004), and (Farley et al., 1995). Each component has a characteristic $^3He/^4He$ value. The anthropogenic ^3He distribution due to the decay of bomb tritium has already been addressed by Ayache et al., 2015.

For this study, we ran two separate simulations, one for each helium component. Each simulation has two boundary conditions: a loss term at the surface, due to the sea-to-air gas exchange, and a source term at the seafloor, describing terrigenic tracer input. Each simulation thus represents the sum of the specified terrigenic component and the atmospheric component, with the distributions of ^3He and ^4He computed separately. We then calculate the isotopic ratio using the δ^3He notation.

3.1.1 Surface boundary condition

The only sink for oceanic helium is loss to the atmosphere. At the air-sea interface, the model will exchange ^3He and ^4He with the atmosphere using sea-air flux boundary conditions that are analogous to those developed for helium during the second phase of OCMIP http://ocmip5.ipsl.jussieu.fr/OCMIP/phase2/simulations/Helium/HOWTO-Helium.html (Dutay et al., 2002). Using the standard flux-gradient formulation for a passive gaseous tracer, the flux of helium, F_{He} is given by:

$$F_{He} = K_w(C_{eq} - C_{surf}) \qquad (2)$$

where K_w is the gas transfer (piston) velocity [m s^{-1}], C_{surf} is the modelled surface ocean concentration of ^3He or ^4He as appropriate, and C_{eq} is the atmospheric solubility equilibrium concentration (Weiss, 1971) at the local sea-surface temperature (SST) and salinity (SSS). Here, we neglect spatio-temporal variations in atmospheric pressure and assume it remains at 1 atm. The gas transfer velocity is computed from surface-level wind speeds, u, [m s^{-1}] from the ARPERA forcing (Herrmann and Somot, 2008; Herrmann et al., 2010) following the Wanninkhof (1992, Eq. 4) formulation:

$$K_w = a\,u^2\,(Sc/660)^{-1/2} \qquad (3)$$

where a = 0.31 and Sc is the Schmidt number which is to be computed from the modelled SST, using the formulation for ^4He given by Wanninkhof (1992), derived from Jähne et al. (1987a). For ^3He, we reduce the Schmidt number (relative to ^4He) by 15% (ScHe-3 = ScHe-4 / 1.15) based on the ratio of the reduced masses, which is consistent with helium isotopic fractionation measurements by Jähne et al. (1987b). Therefore, in the following, the modelled atmospheric component is the helium distribution at equilibrium with surface air-sea boundary conditions, without any helium flux from the seafloor.

3.1.2 Crustal helium fluxes

Lake and groundwater studies have shown that radiogenic helium is continuously released from the underlying crustal bedrock (see Kipfer et al., 2002 for review). Porewaters trapped in oceanic sediments are also enriched in radiogenic ^4He from the underlying oceanic crust and in situ ^4He production by uranium- and thorium-rich minerals, releasing their helium at the sea bottom (Wakita et al., 1985; Sano and Wakita, 1985; Sano et al., 1987; Chaduteau et al., 2009). Deep waters of intra-continental seas such as the Mediterranean are more prone to exhibit a radiogenic ^4He signature than the open ocean because the continental upper crust is about 40 times more enriched in uranium and thorium than the oceanic crust (Taylor and McLennan, 1985; Torgersen, 1989). In the deep eastern Mediterranean, southwest of Crete, extremely high radiogenic ^4He concentrations have indeed been measured in deep brine pools created by the advection of deep buried fluids hosted by the sedimentary matrix beneath the Messinian evaporites (Winckler et al. 1997; Charlou et al., 2003). However, there are no data on the spatial variability of the crustal helium injection to deep waters. Therefore in the model, crustal helium is injected as a uniform flux (in mol of helium per square metre of seafloor > 1000 m) with a $^3He/^4He$ ratio of 0.06 R_{air} (Winckler et al. 1997; Charlou et al., 2003). The initial value of this flux is that estimated by Roether et al. (1998) (Table 2) using a multi-box model in which the thermohaline circulation of the eastern Mediterranean is represented by a deep-water reservoir (> 1000 m depth) and two intermediate water cells (Roether et al., 1994) (see Table 2). Sensitivity tests were made to determine the flux which produces the best agreement with available data (Roether et al., 1998; Roether et al., 2013).

3.1.3 Mantle helium fluxes

The subduction of the African plate below Europe is responsible for the volcanic activity which takes place in the Mediterranean basin (Fig. 3). The main submarine activity is found in the Tyrrhenian and Aegean Seas, and in the Sicily Channel (Dando et al., 1999).

Hydrothermal vents in the Tyrrhenian sub-basin are found all along the Eolian volcanic Arc (Fig. 3) from Palinuro in the north to Eolo and Enarete in the southwest (Lupton et al., 2011), as well as on the Marsili seamount (Lupton et al., 2011).

In the Aegean, hydrothermal systems occur along the southern Aegean Volcanic Arc from Sousaka and Methana in the west to Kos, Yali and Nisiros in the east (Dando et al., 1999).

Finally, a recent helium isotope survey across the Sicily Channel, which separates the Sicilian platform from Africa, also suggests hydrothermal helium input between 600 and 700 m depth associated with the Pantelleria rift (Fourré and Jean-Baptiste, unpublished results).

Location and depth of the active zones are shown in Fig. 3. Table 1 summarizes the ^{3}He fluxes used for our simulations. For the Eolian and Aegean volcanic arc, ^{3}He fluxes were determined by simple scaling to the global ^{3}He flux from arc volcanism, which can be estimated (to within a factor of two) to be ~4×10^{-3} ^{3}He mol per km of arc based on the assumption that the magma production rate of arcs is ~20% of that of Mid-Ocean-Ridges (Torgersen, 1989; Hilton et al., 2002) and the total length of subduction zones. For the Marsili seamount, the ^{3}He flux was estimated from ^{3}He fluxes at nearby subaerial volcanoes (Allard, 1992a, 1992b). ^{3}He/^{4}He isotopic ratios were chosen according to available in situ data (when available) or to ^{3}He/^{4}He data from nearby subaerial volcanoes.

4. Observations used for the comparison with model results

The tracer data in the Mediterranean which are relevant for comparison with model results are the Meteor cruises across the Eastern Mediterranean basin (Roether et al., 2013 - see Fig. 2) and the helium isotope survey carried out by Lupton et al., 2011 in the Tyrrhenian sea. Additional δ^3He data (Fourré and Jean-Baptiste, unpublished data) from the Nov. 2013 Record cruise in the Sicily channel (Geotraces program) are also available. 1987 Meteor section is of particular interest since it is the less affected by tritiugenic ^{3}He (Fig. 2) and therefore the deconvolution of the various helium components using neon is the most accurate. This deconvolution is carried out using the method proposed by Roether et al., 1998; 2001, which allows to derive the atmospheric helium component from the neon distribution and then to obtain the terrigenic helium-4 component by substracting this atmospheric component from the total measured helium concentration. The atmospheric and terrigenic helium-3 components are then obtained using the $^3He/^4He$ ratios of dissolved atmospheric and terrigenic helium, respectively. For the Tyrrhenian sea, the δ^3He excess due to hydrothermal activity along the Aeolian arc is obtained by substracting the background vertical δ^3He profile of vertical cast V01 (see Lupton et al., 2011) to the measured δ^3He. The same method was used for Sicily channel data. Accuracy of the deconvoluted δ^3He is in the range 1%-1.5%.

5. Results

5.1 Crustal helium distribution

We begin our analysis by providing an overview of the simulated crustal+atmospheric helium component. Figure 4a displays a section of modelled $\delta^3He_{crust+atm}$ along a W-E transect across the eastern basin (EMed). As expected, the $\delta^3He_{crust+atm}$ distribution exhibits negative values, predominately in the deep waters, hinting at the presence of crustal-He highly enriched in radiogenic ^4He. The model correctly simulates the crustal-He distribution in the Levantine sub-basin (Fig. 4c), where the simulated $\delta^3He_{crust+atm}$ values agrees reasonably well with observations from Meteor cruise M5. However, modelled $\delta^3He_{crust+atm}$ values for the deep Ionian sub-basin are too low, with a mean value below 3500 m around -7 % compared to –4.5+/-0.7 % in the data (Fig. 4d). This too-large an accumulation of crustal ^4He is the expected consequence of the too-low ventilation of the deep Ionian sub-basin in the model, as already diagnosed in the anthropogenic tritium-^3He simulations of Ayache et al. (2015). The model generates a too-weak formation of Adriatic Deep Waters (AdDW) that prevents the model from reproducing the observed signal associated with injection at depth of surface water.

The simulated $\delta^3He_{crust+atm}$ distribution in the western basin (Fig. 4b) shows the same gradient as in the Levantine basin with negative values in the deep water (values around -5.5%), as a result of the homogenous crustal-He flux over the whole basin (see Sect. 3). In the surface layer helium in solution is essentially in equilibrium with atmospheric helium ($\delta^3He_{crust+atm}$ values around -1.6%), but decreasing steadily with depth down to a layer of minimum $\delta^3He_{crust+atm}$ values in deep waters. Although the terrigenic component cannot been estimated quantitatively for the WMed because of the lack of a precise value for its 3He/4He ratio (R_{ter}), the lower limit of $\delta^3He_{crust+atm}$ (taking R_{ter} equal to zero) is in the range –3.5% - –4.5% for deep waters. This is less radiogenic than in the Eastern basin, in agreement with the conclusions of Rhein et al. (1999) that the crustal component may be small in the WMed. Our model results (-5.5% on average) is somewhat lower, suggesting that, as already observed in the Eastern basin, the model probably underestimates the ventilation rate of deep waters in the western basin too.

5.2 Mantle helium distribution

As discussed above, the main active submarine volcanic systems are located in the Tyrrhenian, Aegean Seas and the Sicily Channel (Fig. 3).

5.2.1 Pantelleria Rift

In the Pantelleria Rift, a clearly visible plume of mantle helium is simulated between 500 and 1000 m depth (Fig. 5a). The modelled δ^3He plume anomaly at 12°5E reaches a maximum value of 2.5% above the atmospheric background of –1.6%. This value is in good agreement with in situ observations at the same location (2.3% above background at 800 m, Fig. 5d; Fourré and Jean-Baptiste, unpublished data).

5.2.2 Tyrrhenian Sea

The submarine volcanic activity in the Tyrrhenian is essentially confined to depths below 1200 m. The corresponding mantle helium input creates a weak but well-defined δ^3He plume (Fig. 5b) centred around 1000 m depth, which propagates into the entire Tyrrhenian sub-basin (Fig. 6). Average simulated δ^3He values above the atmospheric background (-1.6%) are within δ^3He = - 0.5% of the corresponding above-background δ^3He measurements of Lupton et al. (2011) in the same area (Figs. 5b and 5e).

5.2.3 Aegean Sea

Hydrothermal venting in the Aegean sub-basin occurs at shallow depths (between 50 and 450 m depth) compared to the two other sites in the Mediterranean Sea; in consequence the simulated $\delta^3He_{mantle+atm}$ anomaly is particularly weak in this area due to the rapid helium degassing into the atmosphere (Fig. 5c) and the signal does not propagate into the larger area around the Aegean sea (Fig. 6). Note that no δ^3He data are available for comparison in the Aegean basin.

Figure 6 provides a descriptive view of the global distribution of the modelled $\delta^3He_{mantle+atm}$ signal over the Mediterranean Sea. The figure highlights the location of mantle-He sources, and of their propagation through the interior of the Mediterranean Sea. The $\delta^3He_{mantle+atm}$ anomaly is clearly visible over the three main areas of submarine volcanic activity. The mantle-He plume injected by the Aeolian Arc spreads over the entire Tyrrhenian sub-basin, then leaves through the Corsican Channel (1900 m), and extends into the Liguro-Provençal sub-basin associated with the Levantine Intermediate Water (LIW) trajectory, and in the Algerian sub-basin through the Sardinian Channel. The input from the Pantelleria Rift is topographically trapped in the Sicilian channel. The Aegean sub-basin is also impacted by the mantle-He: the He excess is localised in the western part of this sub-basin between mainland Greece and the island of Crete.

5.3 Total helium-3 distribution

The Mediterranean Sea is characterized by coexisting terrigenic and tritiugenic helium throughout its subsurface waters. Fig. 7 presents a model-data comparison of the simulated total δ^3He (sum of terrigenic, tritiugenic and atmospheric helium) in 1987, along the W-E Emed transect corresponding to Meteor 5 cruise (1987). The tritiugenic component in 1987 is taken from Ayache et al. (2015). Figure 7, exhibits a δ^3He maximum at a few hundred metres depth, hinting at the presence of tritiugenic ^{3}He produced by the radioactive decay of anthropogenic bomb-tritium. Further down δ^3He values decrease, and in the Levantine basin, even dropping below the value for solubility equilibrium with the atmosphere (~ -1.6%). This represents the signature of crustal helium in the deep Mediterranean waters.

The model correctly reproduces the δ^3He maximum of the intermediate waters, with values similar to observations, except in the eastern part of the section where it tends to be overestimated. Deeper, we have a realistic simulation of the helium signal in the Levantine sub-basin (Fig. 7b) with δ^3He around -5%, which is in good agreement with observations made during Meteor cruise M5, with only 10% of difference between the simulated δ^3He mean vertical profile and in-situ data below 2000 m depth (Fig. 7b). Again one can clearly see the shortcoming associated with the too-weak EMDW formation in the Adriatic sub-basin, leads to too-negative δ^3He values at depth: the model tends to underestimate the δ^3He levels in the deep water by more than 60 % compared to observations below 2000 m depth (Fig.7c).

Comparison of the tritiugenic and mantle δ^3He signatures, which occur at similar depths in the Mediterranean Sea, shows that tritiugenic ^{3}He clearly dominates over mantle ^{3}He. This finding agrees with those of Roether and Lupton (2011) for the Tyrrhenean basin; they concluded that most of the helium-3 excess is tritiugenic.

6. Discussion

We have presented the first simulation of the terrigenic helium isotope distribution in the Mediterranean Sea, using a high-resolution model (NEMO-MED12). For this simulation we built a source function for terrigenic (crustal and mantle) helium isotopes obtained by simple scaling of published flux estimates (Table 1 and 2). For crustal helium, our helium flux equal to 1.6 10^{-7} mol ^4He m^{-2} yr^{-1}, generates a satisfying agreement with the data in the Levantine basin, where the tritium/^3He simulations of Ayache et al. (2015) have shown that modelled ventilation of the deep waters is correct. This flux represents only 10% of the previous estimate by Roether et al. (1998) for the eastern Mediterranean (1.6 10^{-6} mol m^{-2} yr^{-1}), based on a box-model where the thermohaline circulation of the eastern Mediterranean is represented by a deep-water reservoir (> 1000 m depth) and two intermediate water cells. ~~The Roether et al. (1998) estimate falls in the range of the helium continental flux, 1.4 to 2.2 10-6 mol m-2 yr-1 (see Table 2). However, Winckler et al. (1997) have shown that the thick evaporites layer deposited during the Messinian Salinity Crisisin the Mediterranean Sea acts as a barrier to the upward diffusion of helium from deeper strata. Hence, the expected crustal helium flux from the Mediterranean seafloor may be reduced compared to the "pure" continental value, so the Roether et al. model estimate may be too high.~~

The tritium/^3He (Ayache et al., 2015) and CFC (Palmiéri et al., 2015) simulations have shown that the model adequately represents ventilation of near-surface and intermediate waters but globally underestimates the ventilation rate of the Mediterranean deep waters, particularly in the Ionian sub-basin, where the deep-water ventilation associated with the Adriatic Deep Water (AdDW) is too shallow in the simulations compared to observations. This mismatch is likely due to an overestimation of the freshwater flux (Precipitation-Evaporation and runoff) into the

Adriatic sub-basin. Taking into account this model deficiency, our estimate must definitely be considered as a lower limit of the crustal helium flux into the Mediterranean basin.

For mantle helium, our simple parameterization produces realistic simulated δ^3He values that are in agreement with in situ measurements, thus supporting our scaling approach. This study provides a useful constraint on the magnitude of the hydrothermal helium-3 fluxes in the Mediterranean Sea (Table 1), that is of interest because this flux can be now used to estimate the hydrothermal flux of other chemical species. Hydrothermal venting produces plumes in the ocean that are highly enriched in a variety of chemical species. Hydrothermal activity impacts the global cycling of elements in the ocean (Elderfield and Schultz, 1996), including economically valuable minerals, such as rare-earth elements (REE) which are deposited in deep sea sediments. These minerals are crucial in the manufacture of novel electronic equipment and green-energy technologies (Kato et al., 2011). Hydrothermal chemical elements such as iron also impact biological cycles and eventually the carbon cycle and climate (Tagliabue et al., 2010). Our simulations show that high-resolution oceanic models coupled with measurements of conservative hydrothermal tracers such as helium isotopes can be useful tools to study the environmental impact of hydrothermal activity in a variety of marine environments and at a variety of scales. Beyond the case of hydrothermal activity, it also shows that high-resolution ocean circulation models such as NEMO-MED 12 are well suited to the study of the evolution of quasi-enclosed basins such as the Mediterranean Sea that are under increasing anthropogenic pressure.

The global inventory of helium isotopes in the Mediterranean Sea based on our simulations indicates the relative contribution of each source of the tracer (Table 3). Besides atmospheric helium, which is the main source for both ^3He and ^4He, it shows that tritiugenic ^3He and crustal ^4He are the main contributors to ^3He and ^4He excesses over solubility equilibrium. Therefore, in contrast with the world's oceans where mantle helium dominates over other terrigenic and tritiugenic components, the mantle helium component linked to the submarine volcanic/hydrothermal activity is relatively small compared to the other sources of helium in the Mediterranean Sea. This is due to the cumulated effects of (1) the relatively shallow depths of hydrothermal injections in the Mediterranean (<1000 m) compared to the Mid-Ocean Ridges (MOR), mostly in the range 2000 - 4000 m that favour a more rapid degassing through the air-sea interface; (2) lower helium flux from arc volcanism (20%) compared to MOR volcanism (Torgersen, 1989; Hilton et al, 2002); and (3) high crustal-He flux in the Mediterranean basin due to its intra-continental nature (i.e., with a continental-type crust and high sediment load of continental origin). However, despite its minor contribution to the global helium-3 budget, the hydrothermal component remains identifiable due to its elevated isotopic signature.

7 Conclusions

The terrigenic helium isotope distribution was simulated for the first time in the whole Mediterranean Sea, using a high-resolution model (NEMO-MED12) at one-twelfth of a degree horizontal resolution (6–8 km). The parameterization of the helium injection at the seafloor led to results of sufficient quality to allow us to put valuable constraints on the crustal and mantle helium fluxes. Helium simulations also confirmed some shortcomings of the model dynamics in representing the deep ventilation of the Ionian basin, already pinpointed by recent transient tracer studies. In spite of these limitations and of the limited data set at our disposal for model-data comparison, our work puts additional constraints on the origin of the helium isotopic signature in the Mediterranean Sea. The simulation of this tracer and its comparison with observations provide a new and additional technique for assessing and improving the dynamical regional model NEMO-MED12. This is essential if we are to improve our ability to predict the future evolution of the Mediterranean Sea under the increasing anthropogenic pressure it is suffering (Drobinski et al., 2012). It also offers new opportunities to study chemical element cycling particularly in the context of the increasing amount of data that will result from the international GEOTRACES effort (GEOTRACES, 2007).

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Table 1: Release rates of mantle helium in the Mediterranean Sea used in the model (see§3.1.3).

Region	Prescribed ^{3}He Flux	^{3}He / ^{4}He	References
Tyrrhenian basin:			
Eolian Arc	0.8 (mol yr^{-1})	6 Ra	Sano et al. (1989); Tedesco et al. (1995); Tedesco and Scarsi (1999); Capasso et al. (2005); Capaccioni et al. (2007); Martelli et al. (2008); Fourré et al. (2012)
Marsili Seamount	0.4 (mol yr^{-1})		
Aegean basin:			
South Aegean Arc	1.5 (mol yr^{-1})	4 Ra	Fiebig et al. (2004); Shimizu et al. (2005); D'Alessandro et al. (1997)
Sicily Channel:			
Pantelleria Rift	0.8 (mol yr^{-1})	8 Ra	(Parello et al., 2000)

Table 2 : Release rate of crustal helium used in the model and comparison with crustal helium fluxes in various geological settings.

Region	^3He (mol m^{-2} yr^{-1})	^4He (mol m^{-2} yr^{-1})	References
Mediterranean Sea	1.32×10^{-14}	1.6×10^{-7}	This work
Continental Crust	4.7×10^{-14}	1.4×10^{-6}	(Torgersen, 1989)
Continental Crust	–	2.2×10^{-6}	(Torgersen, 2010)
Eastern Med	–	1.6×10^{-6}	(Roether et al., 1998)
Black Sea	5.8×10^{-13}	0.7×10^{-6}	(Top and Clarke, 1983)
Global Ocean Floor	$(1.5–4.6) \times 10^{-15}$	$(0.2–1.4) \times 10^{-7}$	(Torgersen, 1989)
Pacific Ocean	–	$(0.01–0.2) \times 10^{-7}$	(Sano et al., 1987)
Pacific Ocean	–	0.75×10^{-7}	(Well et al., 2001)

Table 3: Helium inventory (in mole) in the Mediterranean Sea.

	Helium-3	% (Terrigenic)	Helium-4	% (Terrigenic)
Mantle	5	0.8	6.04×10^5	0.3
Crust	18	2.9	2.18×10^8	99.3
Tritugenic (1987)	599	96.3	0	0
Atmospheric	9070		6.67×10^9	
Total	9692		6.89×10^9	

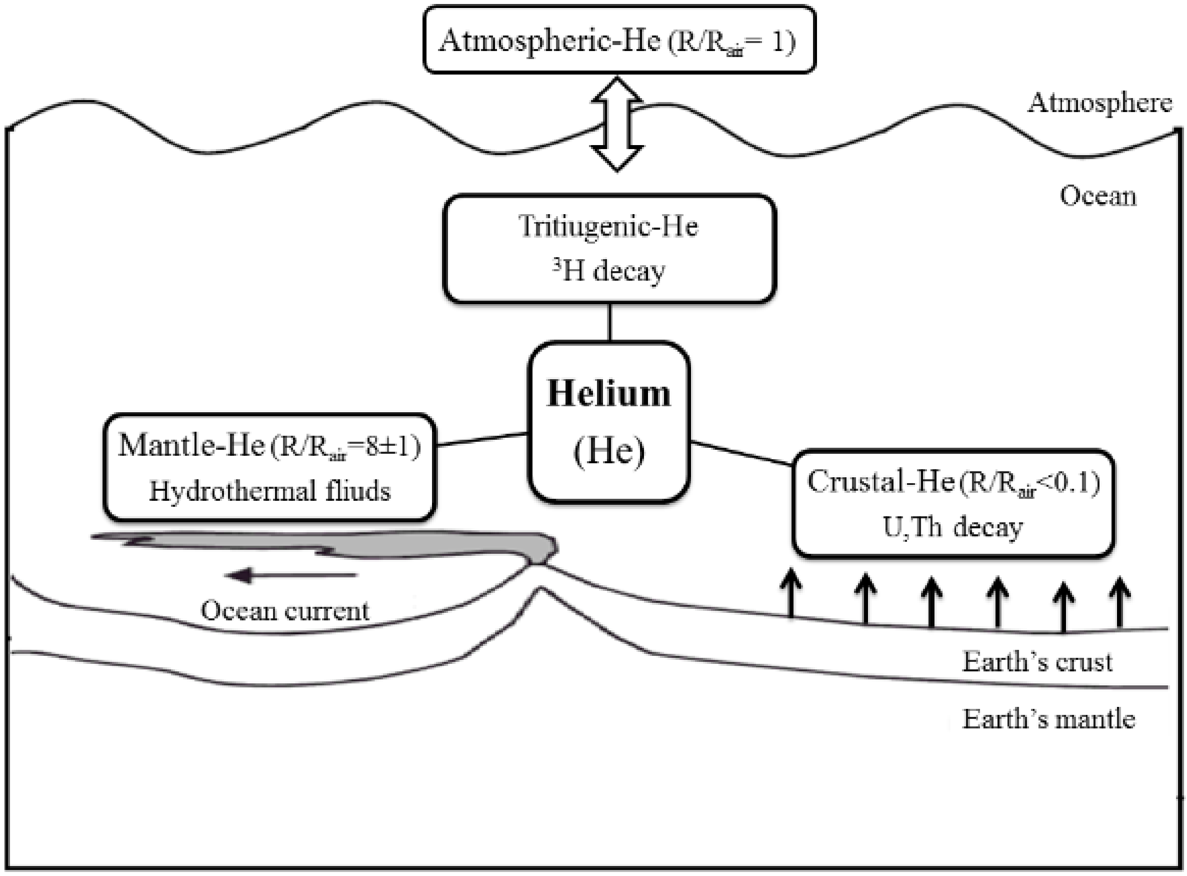

Fig. 1. Schematic of helium components in the ocean. Most of the crustal helium consists of ^{4}He, and most of the mantle helium consists of ^{3}He. Note that the tritiugenic component consists of ^{3}He only. Helium in solution at the ocean surface, is essentially in equilibrium with atmospheric He.

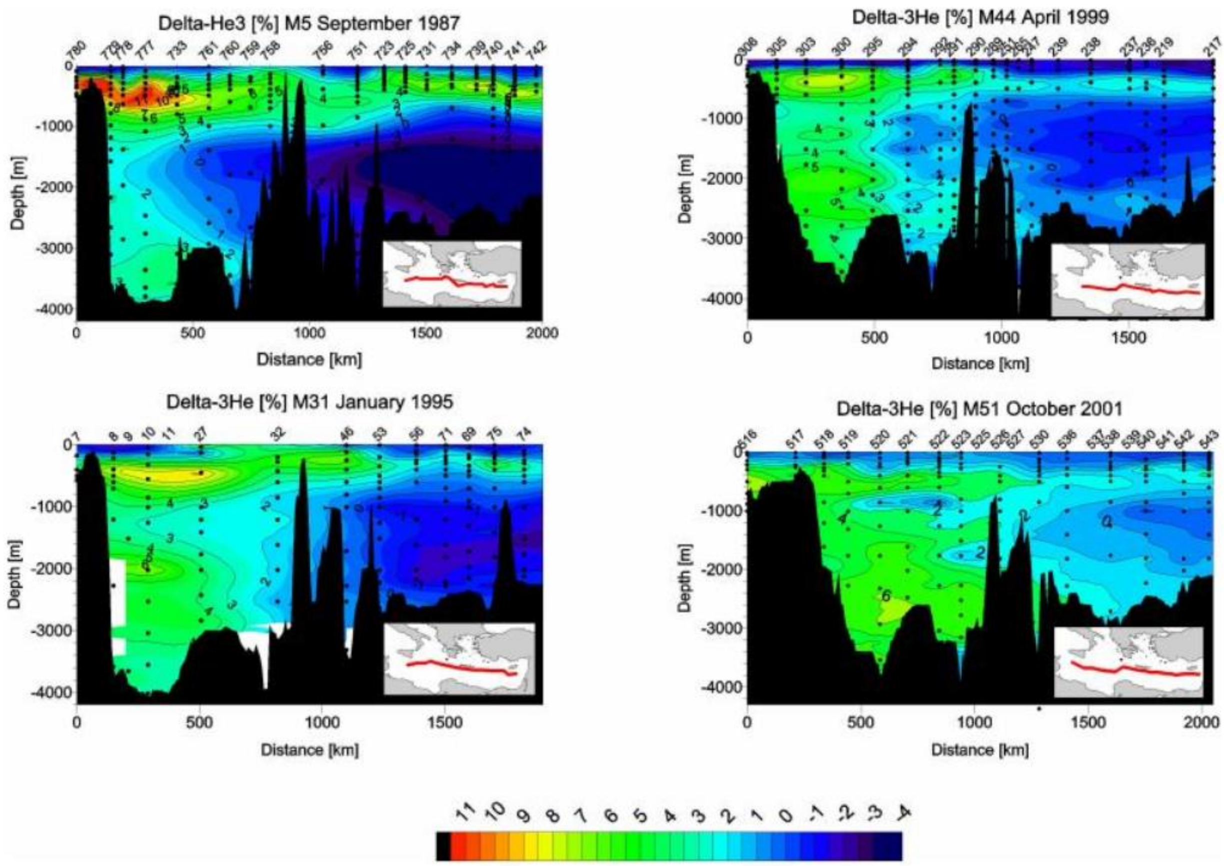

Fig. 2. δ^3He sections of the Meteor cruises in 1987, 1995, 1999 and 2001. Numbers on top are station numbers, observations are indicated by dots, and the actual sections are shown in the inset maps. Isolines are by objective mapping (reproduced from Roether et al., 2013).

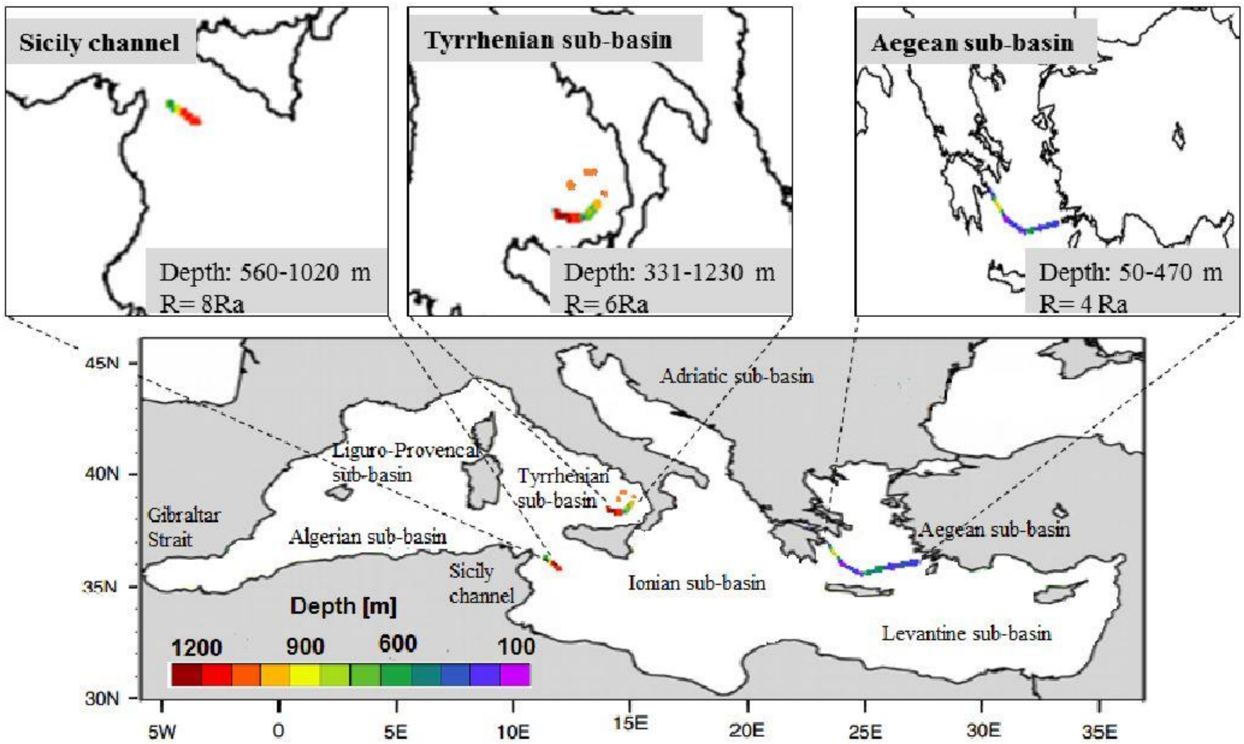

Fig. 3. Depth (in metres) and localization of mantle helium injection in the Mediterranean Sea.

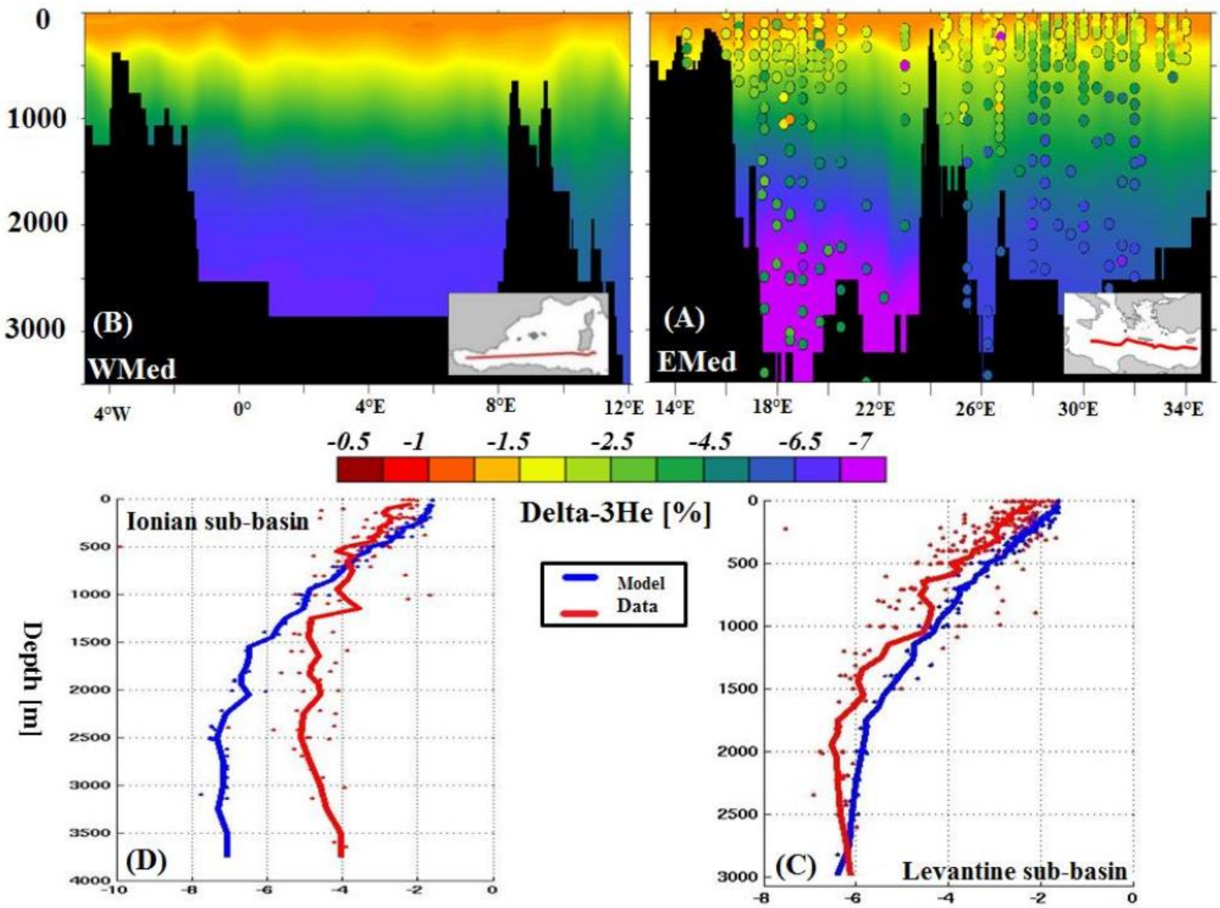

Fig. 4. Crustal+atmospheric δ^3He (in %) model-data comparison along the Meteor M5 (September 1987) section: (a) Colour-filled contours indicate simulated δ^3He (%), whereas colour-filled dots represent the crustal+atmospheric δ^3He deduced from in situ observations using the component separation method of Roether et al., 1998 in the eastern basin (see §4 for details). (b) idem for the western basin (WMed). There are no quantitative data for comparison in the WMed (c) and (d) Comparison of average vertical profiles along the Meteor M5/9-1987 section for the Levantine and the Ionian sub-basins respectively: model results are in blue; red indicates the in situ data.

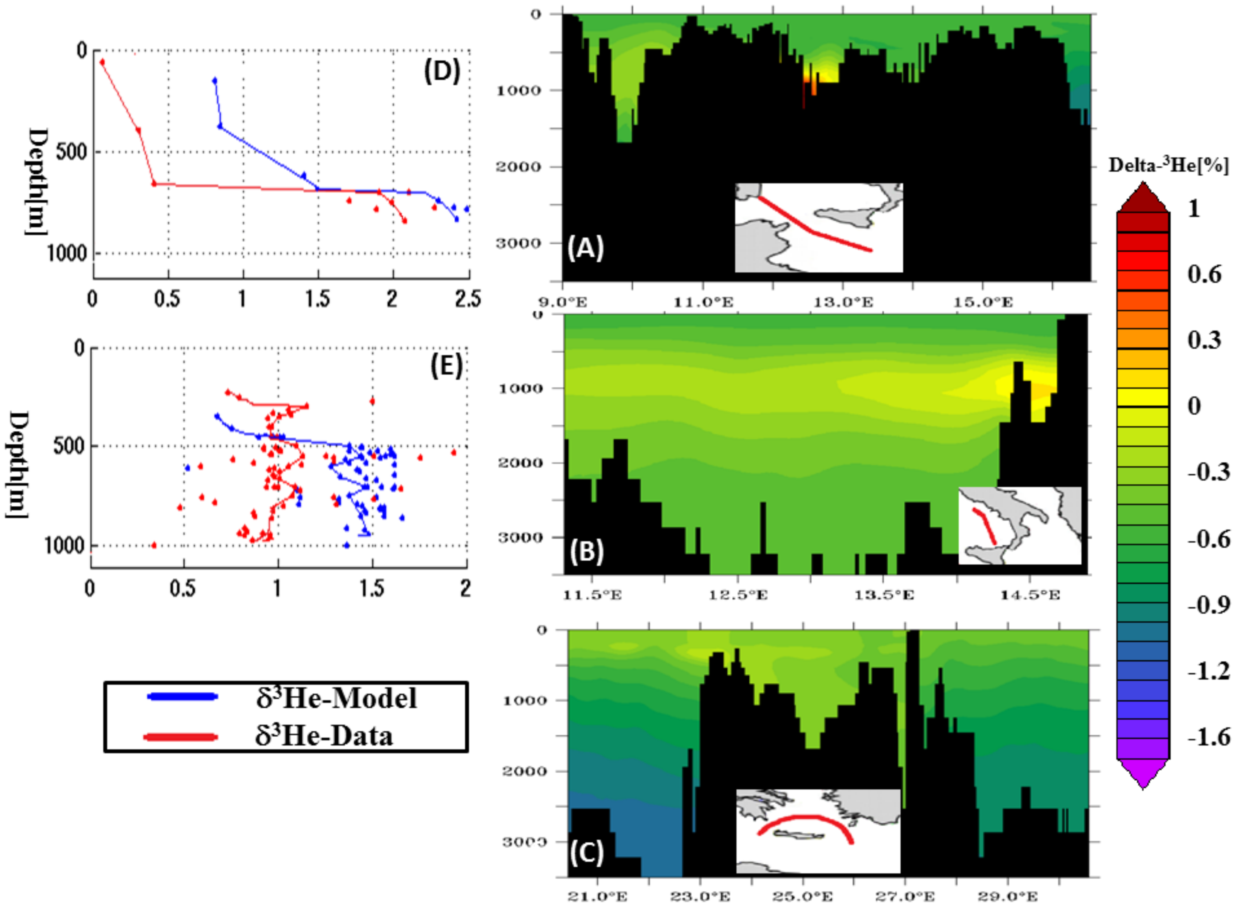

Fig. 5. Mantle+atmospheric δ^3He (%) model-data comparison in (a) the Sicily channel, (b) Tyrrhenian sub-basin, and (c) Aegean sub-basin. (d) Vertical profiles of δ^3He (above the atmospheric background of –1.6%) at 12°5E in the Sicily channel: model results are in blue; red indicates in situ data (Fourré and Jean-Baptiste, unpublished results). (e) Same as (d) for the Tyrrhenian sub-basin. The data are from Lupton et al. (2011). The few stations located right above a plume in Lupton et al. (2011) have been discarded because they cannot be compared to model results which are averaged over the volume of the model cell (~20 km^3). There are no data for the Aegean basin.

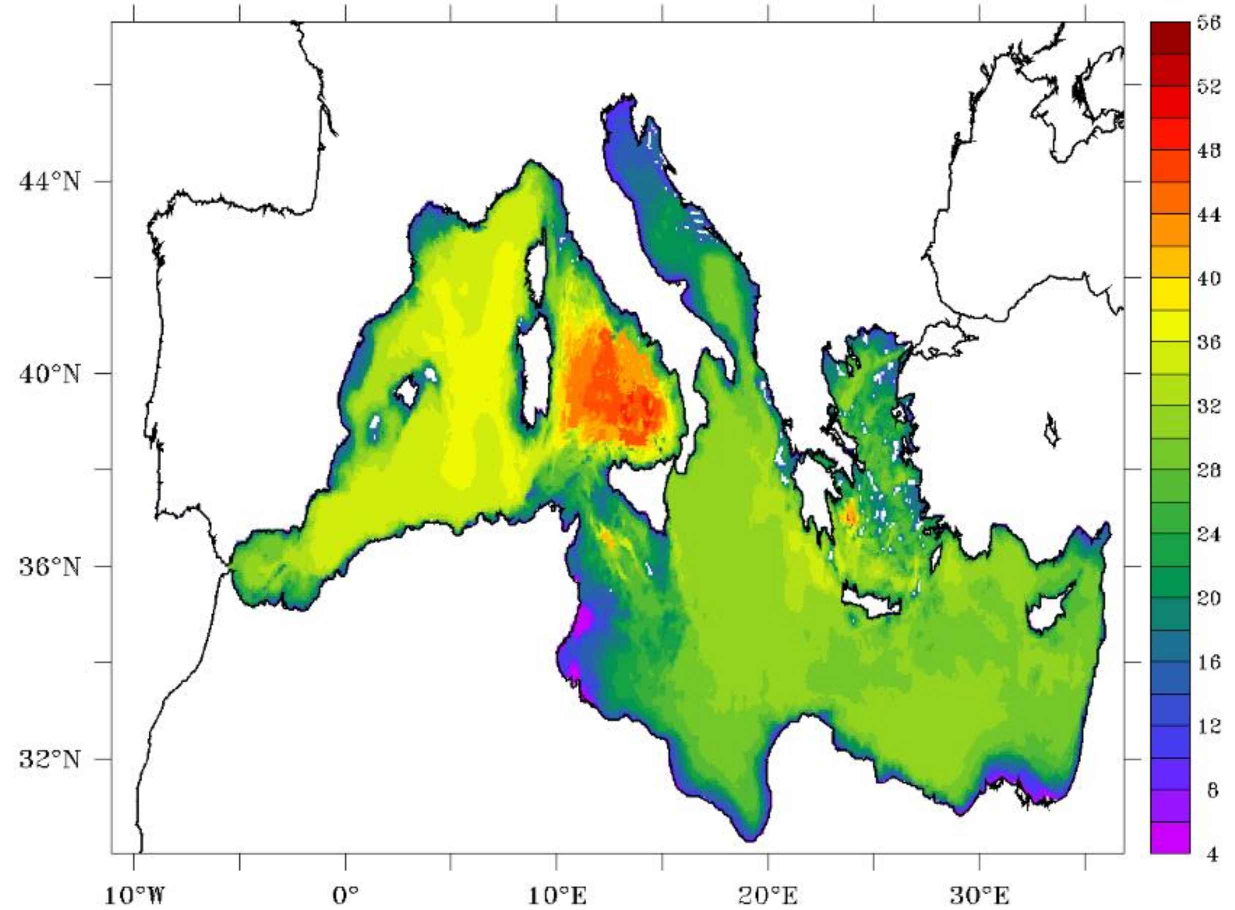

Fig. 6. Horizontal distribution of δ^3He_{mantle} (vertically integrated) across the Mediterranean Sea.

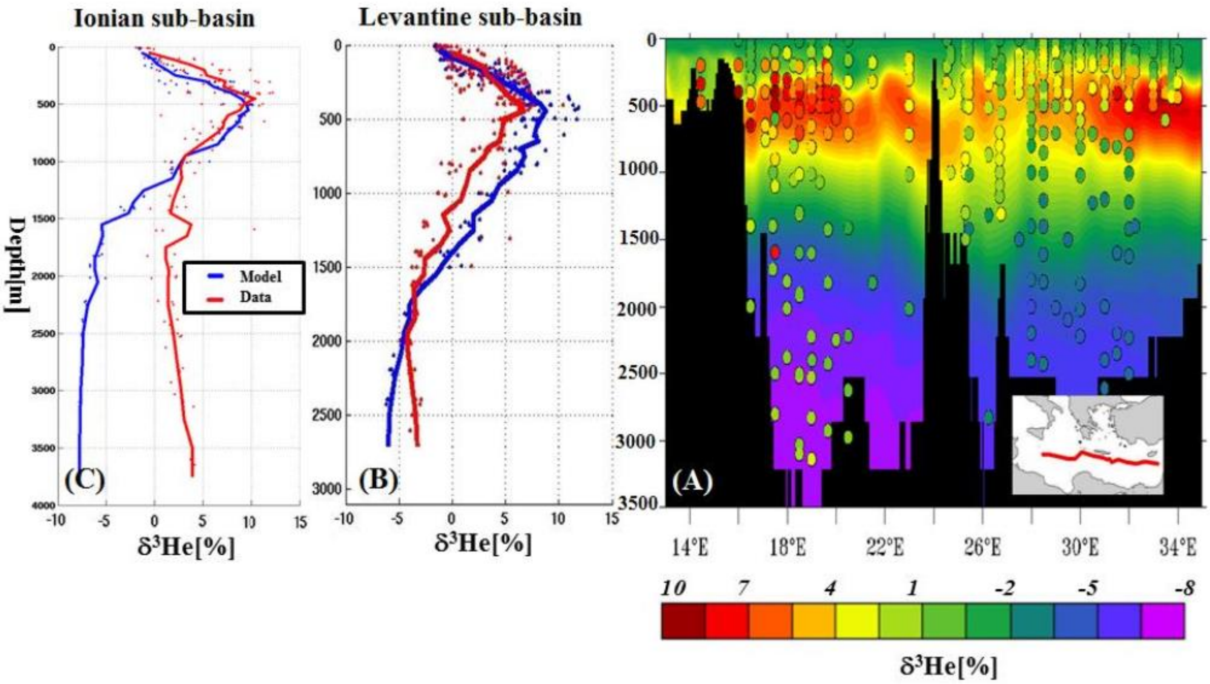

Fig. 7. Total δ^3He (sum of terrigenic, tritiugenic and atmospheric helium) model-data comparison along the Meteor M5 (September 1987) section. (a) Colour-filled contours indicate simulated δ3He (%), whereas colour-filled dots represent in situ observations. (b) and (c) Comparison of average vertical profiles for the Levantine and the Ionian sub-basins respectively. Model results are in blue; red indicates in situ data.