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

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We thank Dr. Matthew Hecht (Editor), Prof. W. Roether and anonymous Referee#2 for their constructive comments and suggestions, which have helped to improve the manuscript. We have carefully considered all questions and concerns raised.

We provide a marked-up manuscript version; all change is recalled in blue (see changes below the reply).

## Reply to Referree#1

This manuscript deals with an interesting modeling topic, worth of publishing in Ocean Science. A high-resolution ocean GCM (NEMO-MED12) is used to simulate the helium isotopes $^3$He and $^4$He in the Mediterranean, distinguishing between the components atmospheric, mantle and crust-derived and tritiugenic $^3$He, comparing the results with observations. The model-data comparison serves to check model performance. The tritiugenic $^3$He is taken from Ayache et. al. (2015), which used the same model. Mantle He is a small contribution, which the authors take from information in the literature.
Rightly, they point out that mantle He in the Mediterranean deep waters is low, because the sources are located at rather shallow depths. The authors conclude that the model simulations are generally realistic, but that the Adriatic source rate and its density are low. The upper boundary condition is He fluxes calculated using established functions of air-sea gas transfer. Their derived crustal He-flux is a factor of 10 lower than obtained in my work Roether et al. (1998). Abstract and Introduction are very good, the English is fine and the list of references is comprehensive. However, I note deficiencies that the authors must consider before submitting their final manuscript

Major items 1. A chapter on the observations used for the comparison, their uncertainties and their treatment is missing.
Response: Done. A specific paragraph has been added to the text (see § 4).

As for differentiating between the components, the text simply mentions (Caption to Fig. 4) that they used the procedures of Roether et al. (1998 and 2013 of which the latter is more advisable;

note that in the former paper the equations are corrupted, the correction paper J. Geophys. Res., 106 (C3),4679 (2001) needs to be consulted).
Response: We used Roether and al.'s methodology but we programmed our own formulas in an Excel spreadsheet. Therefore, our calculations are not affected by the "corrupted" equations.

But that procedure makes use of Ne data, which the authors did not model. It must be clearly specified what procedures were used.
Response: Done. See §4

Fig. 2 shows four Meteor del$^3$He sections, but apparently only the first of them (1987) was used, which must also be stated.
Response: Done. See §4

The choice is natural because the treatment assumes a quasi-steady state circulation. Furthermore, the atmospheric component, which is by far the largest, must be clearly defined, also considering that He solubilities are uncertain by up to 1%.

Response: With respect to data, the atmospheric component is that deduced from measured neon concentrations (see §4). In the model, the atmospheric component is the helium distribution in equilibrium with the atmosphere (= in the absence of any helium flux at sea bottom). This definition has been added at the end of §3.1.1 for the sake of clarity.

2. Apparently the tritiugenic $^3$He results from Ayaches paper earlier this year are used to correct for tritiugenic $^3$He, but In view of the fact that tritiugenic $^3$He dwarfs the terrigenic components (Table 3), I am convinced that the correction lacks the necessary precision. A case in point is Figure 4, which presents simulated and observed data on del$^3$He for the sum of crustal and atmospheric components. The needed correction for tritiugenic $^3$He makes determination of the crustal component rather uncertain.
Response: Tritiugenic $^3$He results from Ayache et al. paper 2013 intervene only in Figure 7 to sum up of all modelled helium components (including tritiugenic $^3$He) for comparison with the delta $^3$He measurements on the 1987 Meteor section. Figure 4, which compares modelled crustal+atmospheric delta $^3$He with the corresponding data, do not require to consider any tritiugenic $^3$He, neither modelled nor deduced from data.

3. Judging from Figure 6, I have the impression that mantle He for the Tyrrhenian is overestimated, although the authors state that they were aware of my paper with John Lupton (OS, 2011) in which we demonstrated that most of the del3He effect is tritiugenic.

Response: As stated in the new §4, mantle delta $^3$He for the Tyrrhenian is calculated by substracting the background delta $^3$He profile of station V01 in Lupton et al (2011) from all measured delta $^3$He vertical profiles. Potential overestimations might have been possible if the stations located right above observed hydrothermal plumes were included in the dataset, but as explained in Fig.5 caption, those stations were discarded.

4. I do not understand how the mantle $^3$He fluxes in Table 1 come about (Section 3.4). For the Tyrrhenian, various authors are cited, but I wonder what their basis was prior to Lupton's $^3$He observations, and to which degree their values are consistent.

Response: As explained in §3.1.3, mantle $^3$He fluxes for the Tyrrhenian (and the Aegean) 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 ~$4\times10^{-3}$ $^3$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. In the absence of data concerning local/regional $^3$He fluxes, this method is the only one at our disposal. Although the method has a large uncertainty (a factor of 2), the reasonable agreement between model and data suggests that this estimate is correct.

I also have doubts about the Sicily Channel values. The text states enhanced $^3$He between 600 and 1000 m depth, which apparently is in the depression in the Sicily Channel. That depression certainly received input by overflow across the eastern ridge by high- del3He waters (mostly tritiugenic) during the early EMT when density was distinctly enhanced (see 1987 section in Fig. 2).

Response: As explained in the new §4, the mantle delta $^3$He in the Sicily channel was calculated in the same way as for the Tyrrhenian (i.e., by substracting the local background delta $^3$He obtained from a station showing no plume-shape $^3$He anomaly). Therefore, any tritiugenic $^3$He contribution might have been removed.

In the last paragraph it is argued on the basis of average release rates of $^3$He as a function of ridge length, for which an uncertainty of a factor of 2 is expected. Might the error not be even higher?

Response: the exact error is difficult to assess but considering that the 20% contribution of arc magmatism to global magmatism is known with an uncertainty of +/- 50% and that the uncertainty on global $^3$He flux at Mid-Ocean ridges is about the same (=50%), the 100% uncertainty on our flux estimate seems reasonable.

5. The discrepancy in the derived crustal He flux density from that in my 1998 paper is tentatively assigned to a possible overestimate in my work. I am convinced, however, that my flux stands on firm ground. The box model that I used was calibrated using observations of CFC-12 and tritium from my 1987 cruise assuming a quasi-steady state situation (Roether and Schlitzer, Dyn Atmosph. Oceans 15, 333-354, 1991). That work gave a renewal time of the Eastern Mediterranean deep waters of about 150 years (a value that never was challenged). This value is the basis on which my 1998 paper converted the 1987 He observations into flux densities of crustal and mantle He (about 5 % mantle He) using literature values for their isotopic composition and assuming steady state (just as assumed in the present work) and an arealy homogeneous mixing. A correction for tritiugenic 3He was made in the deep waters where that correction is small. A 30% uncertainty was reported. With respect to the authors' rate, note that the flux rate naturally adjusts to the vertical transport in the model. The authors admit that the model underestimates the strength and density of the Adriatic source, which after all is the principal deep water source in the eastern Mediterranean. Clearly, thus, the author's value is an underestimate. Because of the mentioned adjustment and considering that the atmospheric component is independent of water turn-over, model- data agreement (Fig. 4) does not prove that the terrigenic flux rate is correct.

Response: Our terrigenic flux is clearly a lower limit because of the weaknesses of the model regarding deep Mediterranean water ventilation rate. We agree that Roether's estimate is much closer to the reality. We corrected the text accordingly to make this clear (removed part in §6).

6. The authors state that the ocean surface He is essentially in solubility equilibrium with the atmosphere (p. 2009, line 10 f.), which means that the limiting step is the net upward transfer of He into the mixed layer from below. I therefore wonder why the authors chose a surface boundary condition in the form of water to air gas exchange (Section 3.2). Having instead assumed quasi-equilibrium at the surface, the vertical tracer gradients in the water column would hardly be different.
Response: we agree that the vertical tracer gradients in the water column would hardly be different. We chose a surface boundary condition in the form of water to air gas exchange to be consistent with the standard protocol used by the model for other trace gases (CFC, SF6 …).

7. p. 2011, line 4 f.: I wonder whether the bottom layer extensions in the model as large as 450 m (p. 2011, line 3 f.) are really suitable (but I am not an expert in this), even if special adjustment to the bottom topography is applied. Especially in the Eastern Mediterranean there are ridges and deep passages that control the deep circulation on vertical scales of less than 100 m. To deal with that is a big challenge for modellers. A further example of such problem is that the EMT-related outflow from the Aegean and its densities obtained by Beuvier et al. (JGR 2010) were low compared with our own assessment in Chapter 6 of
Response: We agree that this is a serious limitation of the model to describe in a fully realistic way the bottom and deep circulation. A new version with higher horizontal and vertical resolutions is being developed, which hopefully will overcome some of the shortcomings in model physics.

8. I note in passing that, had simulated Ne been available, the authors could have obtained a clear separation of the atmospheric component and data on terrigenic $^4$He with no correction for the other He components being needed. Also scale problems in the He data (from measurement, solubility, incomplete equilibration at the surface) could have been avoided.
Response: In constrast with the data, for which neon is essential to separate the various helium components, neon simulations are not necessary to separate the various helium components in the model, since each component is modelled separately.

Technical items

1. P 2009, line 5; A citation for the atmospheric residence time of He is needed.
Done. See §1 line 40

2. p. 2009, line 2 f.: It is stated that the low del$^3$He in the deep layers is erased by the addition of tritiugenic $^3$He. In my view an even larger effect is due to EMT induced upwelling (the T-S correlation was totally changed).

We agree with this remark. A sentence has been added to the text to make this perfectly clear in the revised manuscript. See §1 line 64.
3. p. 2013, line 18: Replace Weiss and Roether (1980) by correct citation (Weiss, 1971?).

Done. See line 163

4. Figure 3: the colors are hard to identify, in an inset showing just the colored lines at higher areal resolution might help.
Done, the revised figure is enlarged.

5. Figure 4, caption: It is stated that data in Western Med to compare with the graph B are missing. Our book chapter mentioned above states, on the basis of the 1997 Poseidon cruise observations, that qualitatively that the crustal component was rather small, with the faster deep water renewal being one possible cause
We thank the referee for this reference. We chose not to use the 1997 Poseidon cruise because, as acknowledge by the referee, the crustal component can only be estimated in a qualitative way due to the lack of a good estimate for the $^3He/^4He$ ratio, $R_{ter}$, of the terrigenic component. However, a lower limit of $\delta^3He_{crust+atm}$ can be estimated (taking $R_{ter}$ equal to zero), which shows that the crustal component is indeed smaller than in the eastern basin, in agreement with Roether's findings. We added a sentence at the end of §5.1 to mention this.

**Reply to Referree#2**

This manuscript presents a high resolution model of the Helium isotopes in the Mediterranean Sea. The authors are using a state of the art model (NEMO) on an area of scientific interest to help bring new knowledge to the scientific community. They offer new values for the Helium isotopes ratio in the Mediterranean Sea which will help modelers of the biogeochemical cycles and the climate better understand the sources of Helium, and constrain the initial conditions for the numerical simulations. Helium studies are useful in the climate simulation community to help describe the ventilation and age of the water masses. While I appreciate the benefit of better constraining these values and command the authors for their work, I think it may be useful to discuss the practical limitations that such work faces when gathering, compiling and synthesizing available data.

The authors use strong words to describe the quality of their findings, which contrasts with the less than optimal datasets they have at their disposal and the practical limitations and simplifications that a modeler has to make when setting up their study.

Response: The conclusion was modified accordingly to stress those limitations (see §7, line 414).

1/ Page 2009: in the delta $^3$Hesw: what does SW mean?

Response: We have used SW for "Sea Water". However, this not necessary in this context so we decided to remove the 'sw' subscript for the sake of clarity.
2/ Page 2009: the value of ratio of $^3He/^4He$ seems intuitive.

Response: All the $^3He/^4He$ values cited in page 2009 are well established values taken from the litterature.

3/Page 2010: discuss a negative ratio.

Response: There are no negative ratios (which would make no sense) but only negative $\delta^3He$. Negative $\delta^3He$, according to its definition as the percentage deviation from the atmospheric ratio, simply means that the $^3$He/$^4$He ratio is lower than the atmospheric ratio. In the ocean, (slightly) negative ratios (around –1.5%) are found in surface waters due to the slightly lower solubility of $^3$He relative to $^4$He, and in some deep waters of intra-continental seas such as the Mediterranean (see introduction) due to the addition of crustal $^4$He from the seafloor and sediment cover.

3a/ Residence time? Ventilation? He from the bomb: distribution linked to circulation: discuss.

Response: The concept of residence time and ventilation are widely used in the oceanographic and tracer community. We added a short definition (with proper references) for those readers not familiar with this (see §1, line 81). Concerning tritiugenic helium (from bomb tritium), we refer to our recent paper (Ayache et al., 2015).

4/Page 2010: Since then helium isotopes... : the authors first refer to a date at which the 3He was discovered then proceed to explain the cycle of the element. "Then" seems to refer to the injection of 3He, not to the time at which it was discovered ($^3$He is being used to trace circulation since it was discovered in 1970 not since it was injected at mid ocean ridges).

Response: This sentence has been changed for the sake of clarity, see §1, line 77.

5/Page 2011: "represent the ventilation of deep waters". The concept of ventilation of water masses should be explained earlier. I think it would help with statements such as that of p 2010 line 1-3.

Response: See response to point n°3a.

6/ The authors alternate the use of "helium" and "$^3$He" throughout the manuscript. Be consistent.

Response: Helium classically designs the sum of both $^3$He and $^4$He isotopes, in practise equal to $^4$He due to the very low isotopic $^3$He/$^4$He ratio in terrestrial samples. He-3 and/or He-4 design the specific isotope which is discussed.

7/ Page 2012: "the exchanges with the Atlantic Ocean are performed through a buffer". I am not familiar with the term buffer used in this context. Rephrase?

Response: NEMOMED12 covers the whole Mediterranean Sea plus a buffer zone including a part of the near Atlantic Ocean (See Figure). The exchanges with the Atlantic Ocean are performed through a buffer zone. From 11°W to 7.5°W, 3D fields relaxed towards in-situ data.

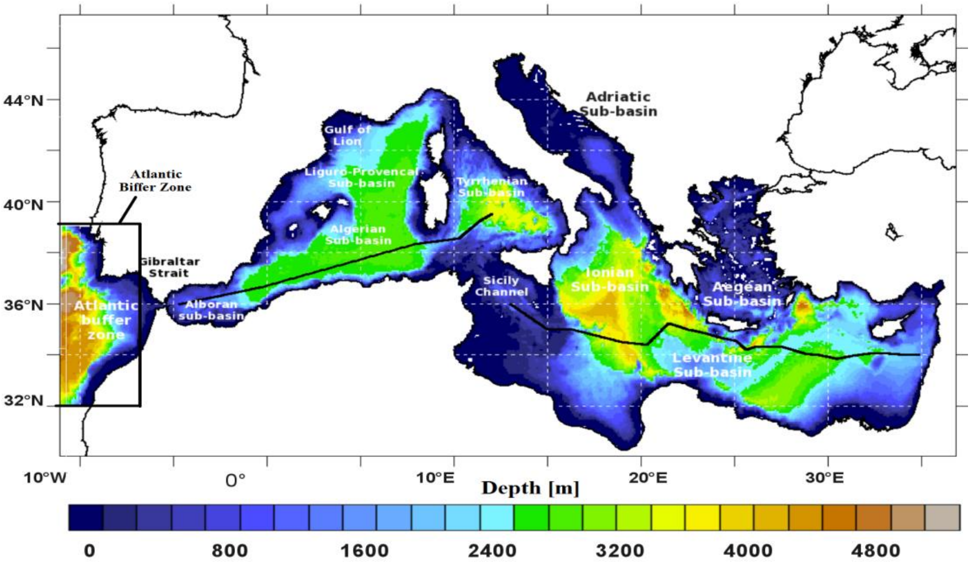

Figure 1. Map of the NEMO-MED12 model domain and bathymetry with location of the main Mediterranean sub-basins. The solid lines represent the trans-Mediterranean sections of the R/V Meteor cruises (used in Fig. 4 and 5).

This sentence has been rephrased in the revised manuscript, (, see §2, line 124).

8/The datasets used in the manuscript cover very different time periods. The temperature and salinity for the Mediterranean sea are prescribed from climatology covering the period 1955-1965. NEMO-MED12 is forced at the surface by ARPERA daily fields of the momentum evaporation and heat fluxes over the period 1958-2013. For the SST a relaxation term is applied to the heat flux. How having 2 different periods for those 2 data source affect the analysis? For the Atlantic buffer the initial state is set from the WOA 2005. How are the possible mismatches in the field values treated?

Response: The physical simulation used here is similar to that described in Beuvier et al. (2012b); Palmiéri et al., (2015); Ayache et al., (2015).
It is initiated in October 1958 with temperature and salinity data representative of the 1955–1965 period using the MEDATLAS dataset (MEDAR/MEDATLAS-Group,2002;Rixen et al.,2005). For the Atlantic buffer, initial conditions are taken from the 2005 World Ocean Atlas for temperature (Locarnini et al.,2006) and salinity (Antonov et al.,2006).
Boundary conditions are also needed to specify physical forcing for the atmosphere, freshwater inputs from rivers and the Black Sea and exchange with the adjacent Atlantic Ocean. For the atmosphere, NEMO-MED12 is forced with daily evaporation, precipitation, radiative and turbulent heat fluxes, and momentum fluxes from the ARPERA data set (Herrmann and Somot , 2008), all over the period 1958–2008. The ARPERA forcing constitutes a 56-year, high-resolution forcing (50 km, daily data) with a good temporal homogeneity (see Herrmann et al.,2010, for more details about the post-2001 period).
To reduce the effect of the initial conditions, we have run a very long spin-up simulation, and we have analysed the outputs only after the steady state situation (after almost 500 years of simulation).

8'/ Page 2014: Each component has a characteristic $^3$He/$^4$He value: can you please elaborate? Or describe the distribution and values so that it is not left to the reader to do so.

Response: For the isotopic characteristics of each component, the reader needs to refer to the introduction and Fig. 1.

9/ Page 2015: Paragraph 3.3: it feels repetitive. It seems that the authors explain the sources of helium repeatedly throughout the paper. While I appreciate the thoroughness of the authors in describing the source mechanism and listing references, I am not sure it is necessary to repeat this throughout the manuscript. Referring the reader to Fig1 cartoon diagram- may be more useful at this point.

Response: this comment is somewhat contradictory with comment 8'/ right above. In paragraph 3.3 we explain why the crustal helium is so important in the Mediterranean Sea, especially for those readers not familiar with helium isotopes.

10/ In the eastern Mediterranean: table 2: why not give the value of the $^{3}$He release rate? Authors list the ratio, and $^{4}$He rate, why not give the $^{3}$He rate?

Response: The $^{3}$He release rate is simply the product of the $^{4}$He release rate multiplied by the $^{3}$He/$^{4}$He ratio. Therefore we feel that an additional column with the $^{3}$He release rate will be somewhat redundant.

11/ Page 2016: typo: needs a "." before "For the Marsili seamount"
Response: Done.

12/ Page 2017. In the 4.1 paragraph. "very similar": well, ... seems to overestimate..

Response: We rephrased this sentence in the revised manuscript, (see §5.1, line 271).

13/ Page 2019: LIW: could you remind the reader what it is?

Response: LIW= Levantine Intermediate Water. This was clarified in the revised manuscript (see §5.3.2, line 320).

14/Page 2019: paragraph 4.3: the notion of "correctly representing" is too vague. The paper would benefit from the use of statistics at this point.

Response: We agree with the referee that a more quantitative analyses would be of interest. Figures 7b and 7c show a comparison of average vertical profiles along Meteor M5 section, which provide quantified estimations of the deviation against observations allowed the identification of the main water masses present in the Med sea (like the Levantine Intermediate Water). Additional quantitative comparison between data and the simulation was added to the text (see §5.3, line 338);

15/ Page 2020: typo: Crisisin?

Response: Done.

16/ Page 2023: "It 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." While I do understand and agree with this statement, are NEMO simulations coupled to a real atmospheric model? it seems to be that it is a bit difficult to do ocean only simulations for climate modelling purposes.

Response: NEMO is the oceanic component of the regional modelling platform MORCEMED (Model of the Regional Coupled Earth system) focusing on the Mediterranean basin. Based on coupling of existing regional models of the various components of the Earth system (ocean, continental land masses, atmospheric composition) and interfacing with the IPSL's global climate model to study the evolution of the Mediterranean sea under the increasing anthropogenic pressure (Drobinski et al., 2012). (See §7, line 420.).

17/ Figure 2 caption: remind the reader which area the Meteor Cruise looks at, as there are not lat/long reference on the figure.

Response: As indicated in the caption of Fig.2, the location of the Meteor sections are shown in the inset maps.

18/ Figure 4 caption: there is a typo: double "the".

Response: Done

There is no explanation about how the straight lines are obtained from the dotted clouds on subfig C/ and D/.

Response: Fig.4c and 4d: As indicated in the caption, the straight lines represent the average of all individual measured or modelled points (represented by the “dotted clouds”).

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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, $R_{air} = {}^3He/{}^4He = 1.384 \times 10^{-6}$ (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 $^3$He/$^4$He variations are usually expressed as $\delta^3$He, the percentage deviation from the atmospheric ratio, defined as $(R_{sample} / R_{air} - 1)100$. Below the mixed layer, oceanic $^3$He/$^4$He values are usually significantly higher than the atmospheric ratio, with $\delta^3$He 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 $^3$He/$^4$He ratios indicative of a preferential addition of $^4$He-rich crustal helium rather than $^3$He-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 $\delta^3$He 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 $^3$He 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); $U.\nabla C$ is advection of the tracer along the three perpendicular axes and $\nabla.(K\nabla C)$ is the lateral and vertical diffusion, with the same parameterization as for the hydrographic tracers.

Because $^3$He, $^4$He 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 $^{3}$He and $^{4}$He 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 $10^{-2}$ $\delta^{3}$He % 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 $^{3}$He/$^{4}$He value. The anthropogenic $^{3}$He 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 $^{3}$He and $^{4}$He computed separately. We then calculate the isotopic ratio using the $\delta^{3}$He 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 $^3$He and $^4$He 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 $^3$He or $^4$He 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 $^4$He given by Wanninkhof (1992), derived from Jähne et al. (1987a). For $^3$He, we reduce the Schmidt number (relative to $^4$He) 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 $^4$He from the underlying oceanic crust and in situ $^4$He 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 $^4$He 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 $^4$He 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 $^3$He/$^4$He 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 ~$4\times10^{-3}$ $^{3}$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 $\delta^{3}$He 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 $^3$He/$^4$He ratios of dissolved atmospheric and terrigenic helium, respectively. For the Tyrrhenian sea, the $\delta^3$He excess due to hydrothermal activity along the Aeolian arc is obtained by substracting the background vertical $\delta^3$He profile of vertical cast V01 (see Lupton et al., 2011) to the measured $\delta^3$He. The same method was used for Sicily channel data. Accuracy of the deconvoluted $\delta^3$He 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 $^4$He. 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 $^4$He 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-$^3$He 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 $\delta^3$He 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 $\delta^3$He plume (Fig. 5b) centred around 1000 m depth, which propagates into the entire Tyrrhenian sub-basin (Fig. 6). Average simulated $\delta^3$He values above the atmospheric background (-1.6%) are within $\delta^3$He = - 0.5% of the corresponding above-background $\delta^3$He 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 $\delta^3$He 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 $\delta^3$He (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 $\delta^3$He 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 $\delta^3$He 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 $\delta^3$He 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 $\delta^3$He around -5%, which is in good agreement with observations made during Meteor cruise M5, with only 10% of difference between the simulated $\delta^3$He 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 $\delta^3$He values at depth: the model tends to underestimate the $\delta^3$He levels in the deep water by more than 60 % compared to observations below 2000 m depth (Fig.7c).

Comparison of the tritiugenic and mantle $\delta^3$He 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 $^4$He $m^{-2}$ $yr^{-1}$, generates a satisfying agreement with the data in the Levantine basin, where the tritium/$^3$He 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/$^3$He (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 $\delta^3$He 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 $^3$He and $^4$He, it shows that tritiugenic $^3$He and crustal $^4$He are the main contributors to $^3$He and $^4$He 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 | $^3$He (mol m$^{-2}$ yr$^{-1}$) | $^4$He (mol m$^{-2}$ yr$^{-1}$) | References |
|---|---|---|---|
| Mediterranean Sea | $1.32 \times 10^{-14}$ | $1.6 \times 10^{-7}$ | This work |
| Continental Crust | $4.7 \times 10^{-14}$ | $1.4 \times 10^{-6}$ | (Torgersen, 1989) |
| Continental Crust | – | $2.2 \times 10^{-6}$ | (Torgersen, 2010) |
| Eastern Med | – | $1.6 \times 10^{-6}$ | (Roether et al., 1998) |
| Black Sea | $5.8 \times 10^{-13}$ | $0.7 \times 10^{-6}$ | (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 | – | $0.75 \times 10^{-7}$ | (Well et al., 2001) |

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

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

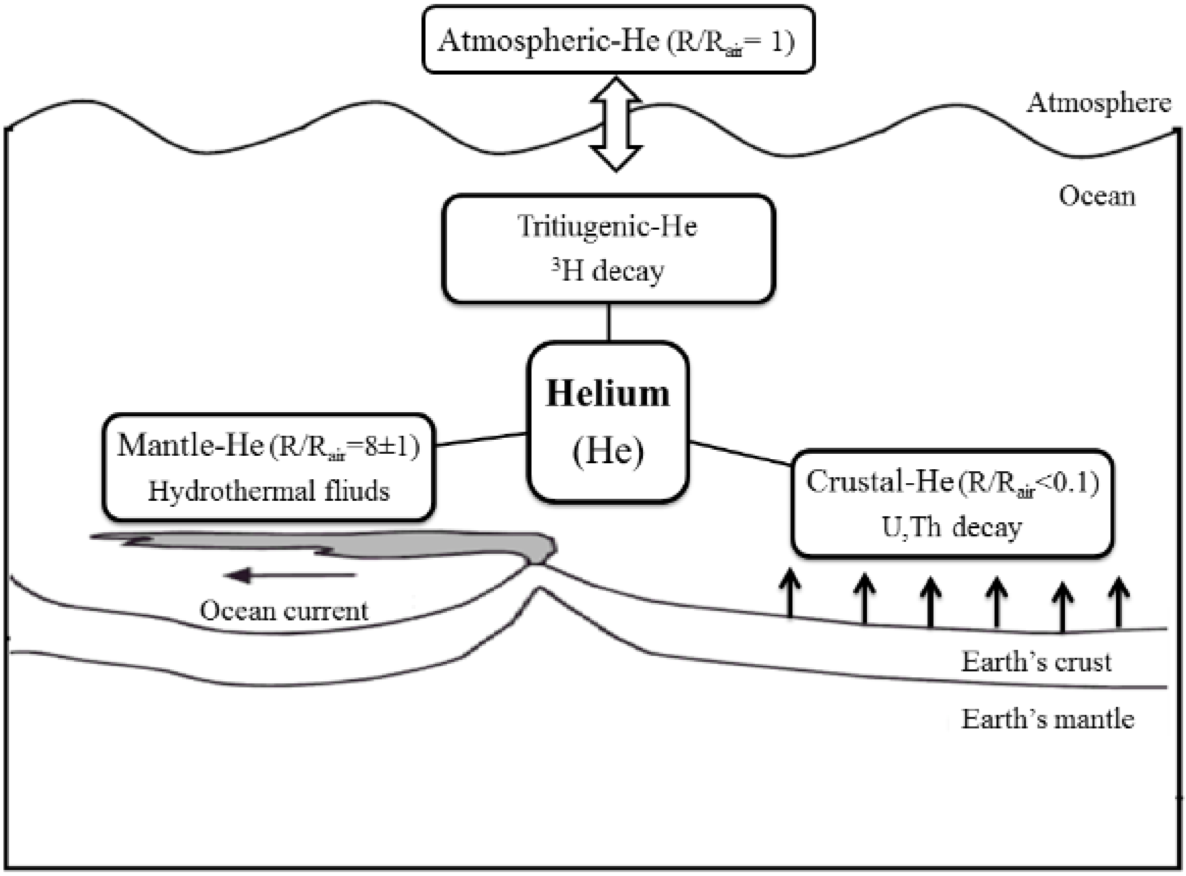

**Fig. 1**. Schematic of helium components in the ocean. Most of the crustal helium consists of $^4$He, and most of the mantle helium consists of $^3$He. Note that the tritiugenic component consists of $^3$He only. Helium in solution at the ocean surface, is essentially in equilibrium with atmospheric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

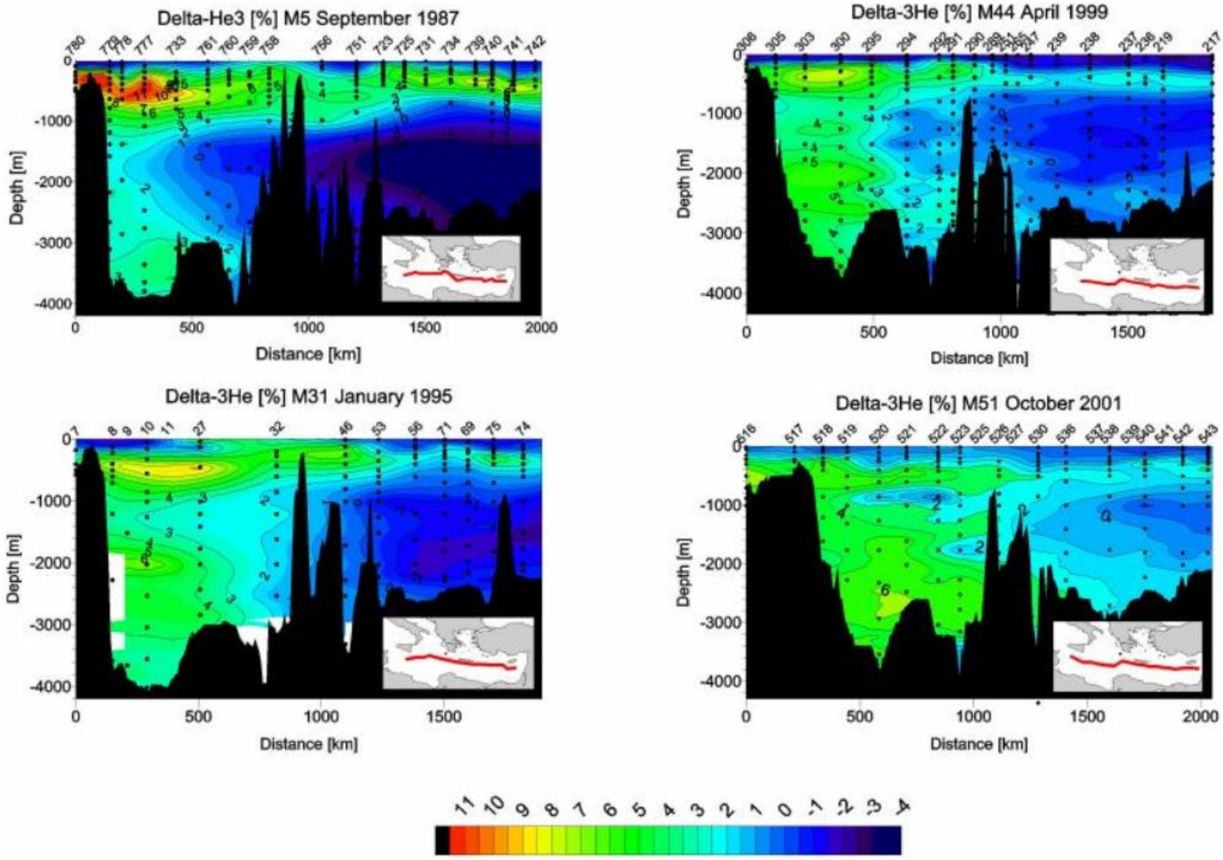

**Fig. 2.** $\delta^3$He sections of the Meteor cruises in 1987, 1995, 1999 and 2001. Numbers on top are station numbers, observations are indicated by dots, and the actual sections are shown in the inset maps. Isolines are by objective mapping (reproduced from Roether et al., 2013).

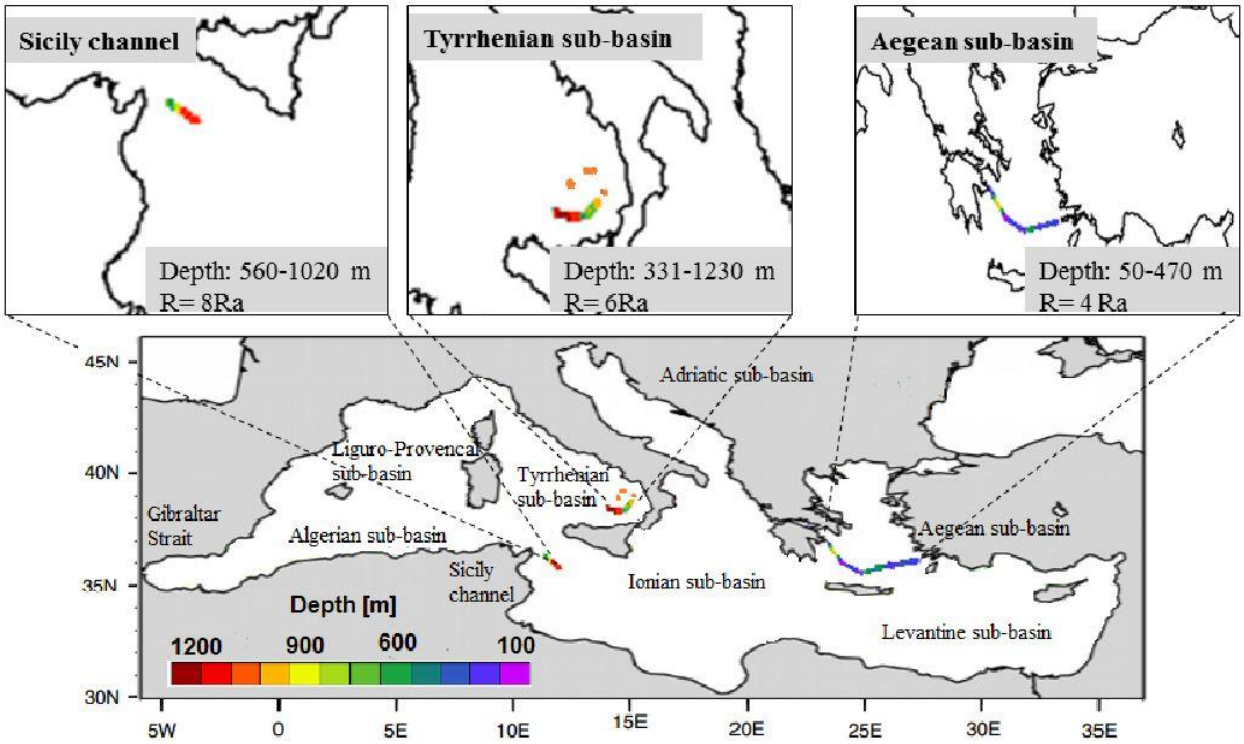

**Fig. 3.** Depth (in metres) and localization of mantle helium injection in the Mediterranean Sea.

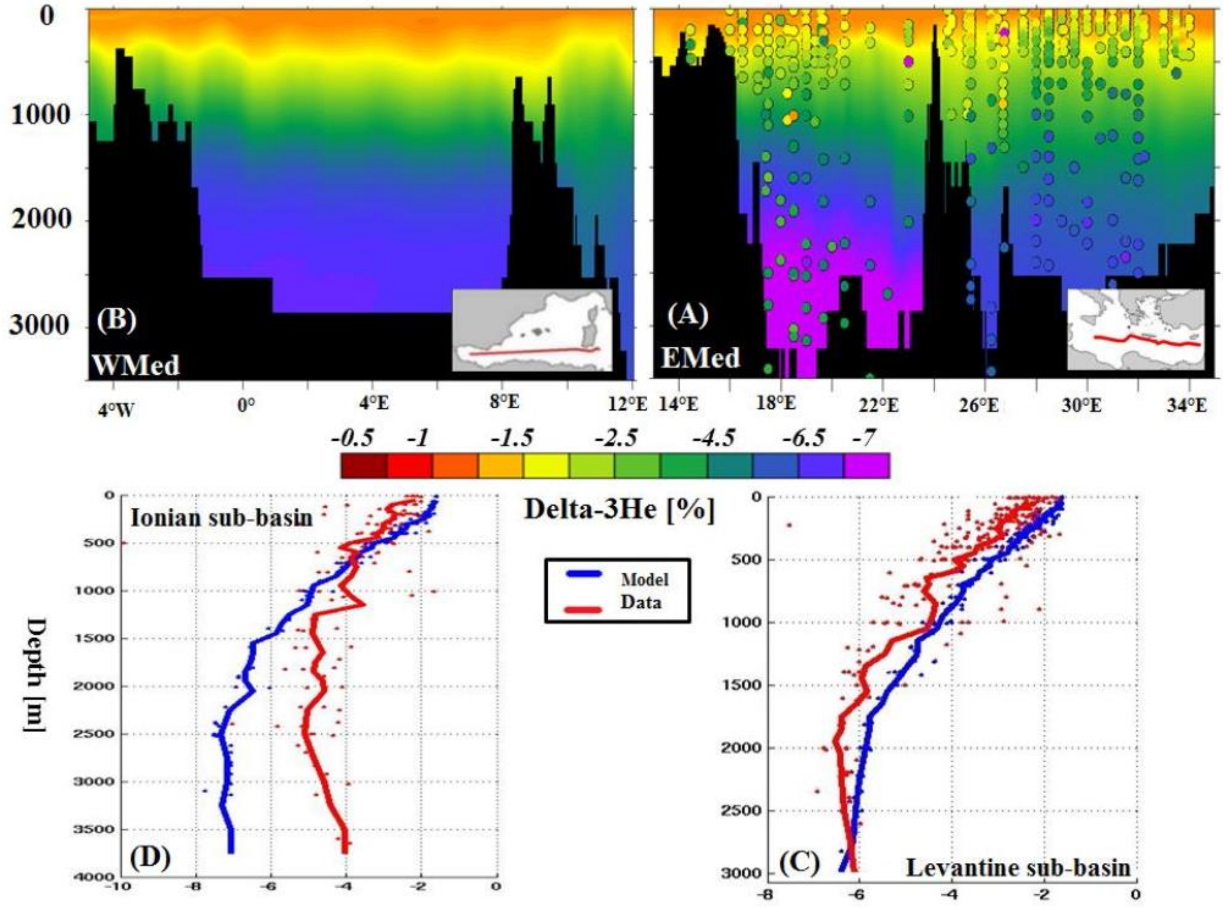

**Fig. 4**. Crustal+atmospheric $\delta^3$He (in %) model-data comparison along the Meteor M5 (September 1987) section: (a) Colour-filled contours indicate simulated $\delta^3$He (%), whereas colour-filled dots represent the crustal+atmospheric $\delta^3$He deduced from in situ observations using the 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.

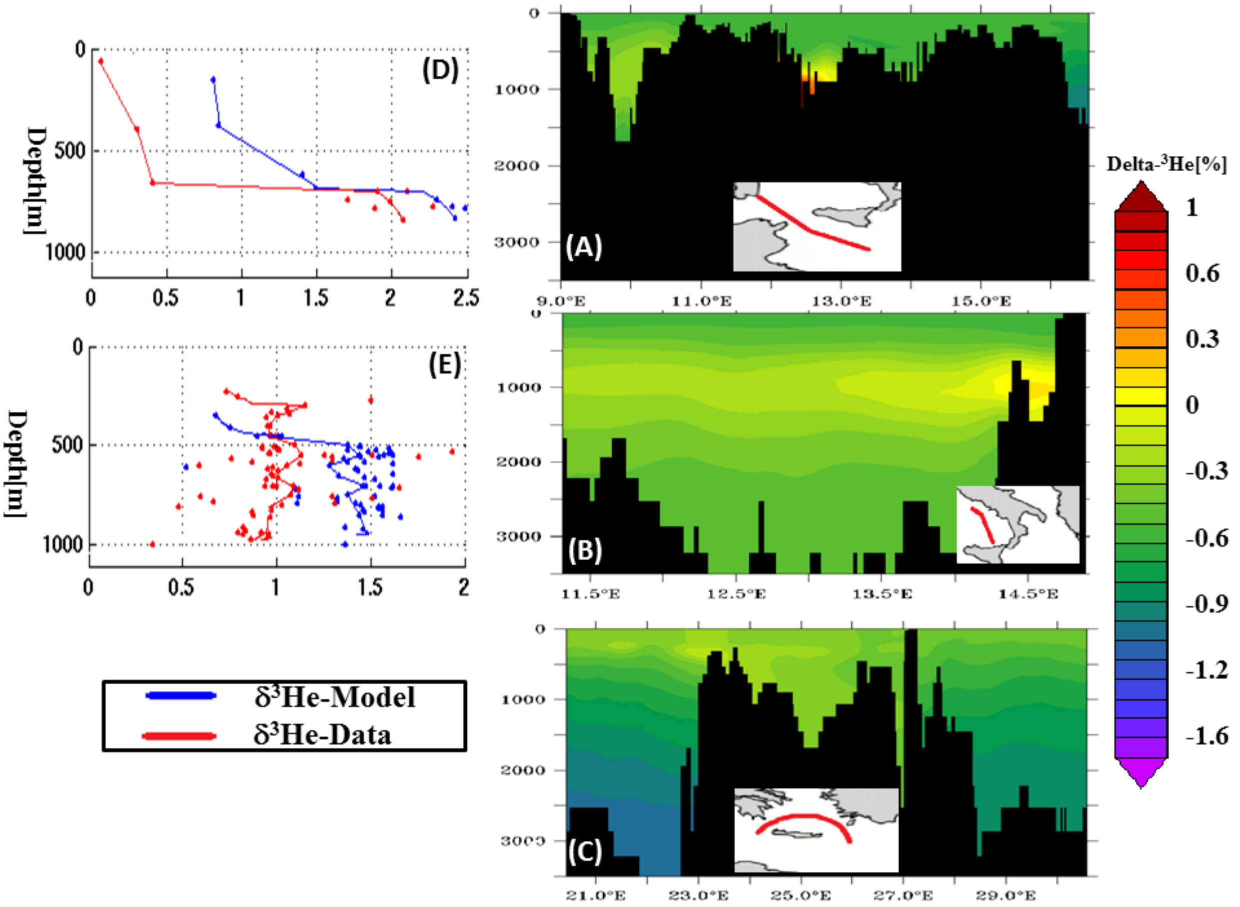

**Fig. 5.** Mantle+atmospheric $\delta^3$He (%) model-data comparison in (a) the Sicily channel, (b) Tyrrhenian sub-basin, and (c) Aegean sub-basin. (d) Vertical profiles of $\delta^3$He (above the atmospheric background of –1.6%) at 12°5E in the Sicily channel: model results are in blue; red indicates 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.

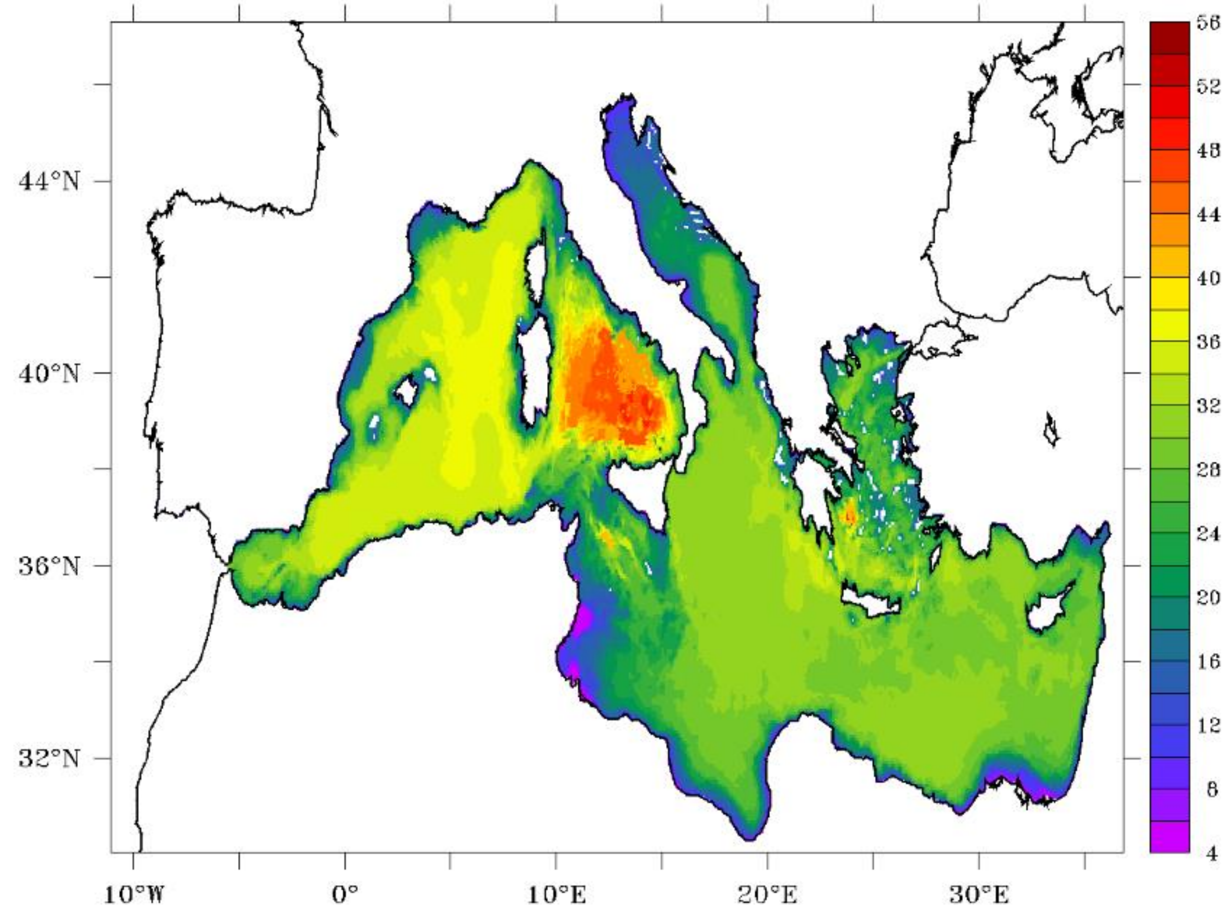

**Fig. 6.** Horizontal distribution of $\delta^3He_{mantle}$ (vertically integrated) across the Mediterranean Sea.

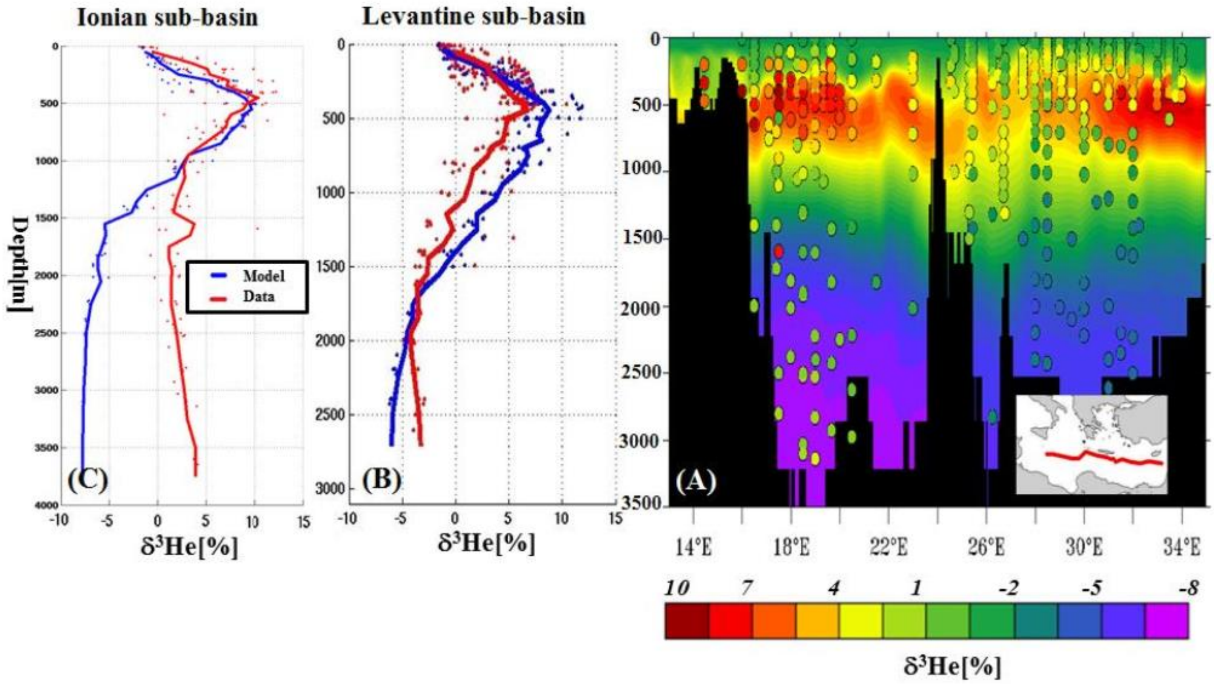

**Fig. 7**. Total $\delta^3$He (sum of terrigenic, tritiugenic and atmospheric helium) model-data comparison along the Meteor M5 (September 1987) section. (a) Colour-filled contours indicate simulated $\delta$3He (%), whereas colour-filled dots represent in situ observations. (b) and (c) Comparison of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