The author's respond to the Reviewer #1

We are very grateful to the Reviewer for a very careful reading of the manuscript and a number of useful comments and critics we tried to take into account in the revised version. Our point-to-point responses can be found below.

The main changes made

We have modified our simulation code and recalculated all the results with this new code. The code, used in the previous version of our manuscript (ms) for plotting figures, underestimated the amount of yellow tracers from the area around the Fukushima Nuclear Power Plant (FNPP) advected outside that area and had a high risk to be contaminated. As before, we distribute a large number of particles in a large area in the northwestern Pacific on a fixed date and advected them backward in time. In the previous code, the particles, which crossed the yellow rectangular around the FNPP (Fig.1a) in the past for the period from the day of accident, March 11, 2011 to May 18, 2011, have been marked by the yellow color on the corresponding Lagrangian map. The particles, which were present in that area and leaved it after May 18, have not been colored in yellow. However, those particles also have a risk to be highly contaminated and should be specified as yellow ones. The present code specifies all those particles as yellow ones. As the result, some "white" waters, which have not been specified previously, now have been specified to came from the yellow area around the FNPP with a high risk to be contaminated.

We've cardinally rewritten Secs. 3.1, 3.2 and 3.3 to compare more clearly our simulation results with the measurements by Buesseler2012, Kaeriyama2013, Kuramoto2014 and Budyansky2015. With this aim we imposed on Figs.2c, 2d, 4a and 4d locations of stations with measured values of the cesium concentration levels in collected surface seawater samples in 2011 and 2012.

When working on a revised version of our ms, we have found the paper by Kumamoto, Y. et al. Southward spreading of the Fukushima-derived radiocesium across the Kuroshio Extension in the North Pacific. Sci. Rep. 4, 4276; DOI:10.1038/srep04276 (2014). Seawater samples for radiocesium measurements in the frontal area have been collected during the R/V 'Mirai' cruise in the very beginning of February 2012. We used this new possibility to compare our simulations with this new data. We imposed on the simulated Lagrangian map in Fig.4a locations of stations to the north of the Kuroshio Extention (>36N) with measured levels of the cesium concentrations and found a good qualitative correspondence of those measurements with our simulation results 10 months after the accident in the sense that stations with measured background level are in the area of Oyashio ``blue" waters with low risk to be contaminated, whereas stations with comparatively high level of radiocesium concentrations are in the area of Fukushima-derived ``yellow" waters with increased risk of contamination.

I. Reviewer #1: Recommendation -

This paper presents Lagrangian maps that visualize the origin, history and fate of the water masses in the mesoscale eddies off the coast of Japan. From the results, the authors argued: 1) the potential risk of contaminated water derived from the Fukushima nuclear power plant, 2) transition of water properties included in the mesoscale eddies and 3) qualitative correspondence between the Lagrangian maps with the observed Cs-137 data (Buesseler et al., 2012, Kaeriyama et al., 2013).

The methodology is very interesting and the authors arguments 1) and 2) are understandable. However, the argument 3) is not supported by the results presented by this work. Consequently, I recommend to reinforce the argument 3) with their results presented in this work. The concrete problems regarding the argument 3) are described below.

Responses to the First Reviewer's report (Problems regarding the argument 3) 1. Cited from the referee's report

p. 9 L. 32-p. 10 L. 7: This paragraph is totally describing results by Prants et al. (2014) and

not by this paper. The observed data by Kaeriyama et al. (2013) showed especially high Cs-137 concentrations in the green segment of Fig. 2d. The result of this study showed that the green segment is corresponding to the blue color water mass (Fig.2d), which means Cs-137 concentrations are low. The authors need to address this inconsistence and discuss possible reasons explaining it.

Our response

We removed that paragraph from the revised test. We recalculated all the results and imposed on the updated version of Fig. 2d locations of stations in the end of July 2011 with measured values of the cesium concentration levels in collected surface seawater samples by Kaeriyama et al. (2013). We removed colored segments in updated version of Fig.2. The green segment in former Fig. 2d corresponds to stations 43 (41N), 44 (40.5N), 45 (40N) and 46 (39.5N) marked by the magenta diamonds in the updated version of Fig. 2d.

'Station 43 was located inside the anticyclone HE filled mainly by ``yellow'' waters, and we estimate the risk to found Fukushima-derived radionuclides there to be large. Station 44 was located at the southern periphery of the anticyclone HE at the boundary between ``white'' and ``blue'' waters but in close proximity to a ``yellow'' streamer.' (cited from the updated version of Sec.3.2).

As to stations 45 and 46, the following text has been added to the updated version of Sec.3.1:

'However, there is an inconsistence of simulation with samplings at stations 45 and 46 where there are practically now yellow tracers but only blue and white ones. The reasons of this inconsistence might be different. In this paper we track only those tracers which were originated from the blue, red and black segments and the yellow rectangular around the FNPP shown in Fig.1a. So we did not specify the origin of white waters. They could reach their places on the maps from anywhere besides those segments and the area around the FNPP. They could in principle contain Fukushima-derived radionuclides deposited at the sea surface from the atmosphere after the accident and then being advected by eddies and currents in the area. Moreover, they could be those tracers which have been located inside AVISO grid cells near the coast around the FNPP just after the accident and then have been advected outside. We removed from consideration all the tracers entered into any AVISO grid cell with two or more corners touching the land because of inaccuracy of the altimetry-based velocity field there and in order to avoid artifacts. Thus, the white streamers inside the core and at the periphery of the blue cyclone with the center at $N{39.7}$, $E{144.2}$ at the places or nearby stations 45 and 46 with high measured concentrations of cesium by \citep{Kaeriyama13}, could, in principle, contain contaminated water. However, it has not been proved in our simulation by the mentioned reasons' (cited from the updated version of Sec.3.1).

2. Cited from the referee's report

p. 10 L. 26-29: Kaeriyama's transect is not reaching to "yellow water"; I do not agree with the authors conclusion is supported by the observation by Kaeriyama et al. (2013).

Our response

As it is clearly seen in the updated version of Fig. 2d, the Kaeriyama's transect reaches to 'yellow' water. Its northern station 43 was located inside the anticyclone HE filled mainly by `yellow" waters, and we estimate the risk to found Fukushima-derived radionuclides there to be large. It is supported by the measurements of concentration of Cs 137 by Kaeriyama et al. (2013) to be around 71 mBq/kg at that station.

3. Cited from the referee's report

p. 11 L. 4-13: This paragraph is describing results by Budyansky et al. (2015). There are no discussion for correspondence between the Lagrangian map with the observed data.

Our response

Our ms is intended for a wide audience, not only to specialists in radioactivity measurements. The corresponding paragraph really describes mainly some results by Budyansky et al. (2015). We put it to the context because the measurements by Budyansky et al. (2015) and some simulation results in that paper support our conclusions about the role of the Tsugaru eddy (TsE) in transport and mixing of Fukushima-derived radionuclides. Moreover, we imposed on updated version of Fig. 4d location of station 84 with measured increased values of the cesium concentration levels in July2012, 15 months after the accident.

Other isuues

4. Cited from the referee's report

p. 9 L. 11: The meridional transect by Buesseler et al. (2012) is 144E, not 145E.

Our response

Thank you, it's our mistake. The meridional transect by Buesseler et al. (2012) is shown in Fig.2c and in its caption to be along 144E, not 145E. We corrected this mistake in the main text.

5. Cited from the referee's report

It is strongly recommended to add figures comparing the Lagrangian maps with the observed data. The comparisons are described in text, but they are hard to understand as readers need to look around the papers Buesseler et al. (2012) and Kaeriyama et al. (2013). Their data are publicly available and number of the data are not so many, it is easy to make figures to compare the Laglangian maps with the observed data.

Our response

We have done that and imposed on Figs.2c, 2d and 4d locations of stations with measured values of the cesium concentration levels in collected surface seawater samples in 2011 and 2012. We thank again the Reviewer for advising that. It helped us not only to improve the ms but forced us to recalculate all the results.

The author's respond to the Reviewer #2

We are very grateful to the Reviewer for a very careful reading of the manuscript and a number of useful comments and critics we tried to take into account in the revised version. Our point-to-point responses can be found.

The main changes made

We have modified our simulation code and recalculated all the results with this new code. The code, used in the previous version of our manuscript (ms) for plotting figures, underestimated the amount of yellow tracers from the area around the Fukushima Nuclear Power Plant (FNPP) advected outside that area and had a high risk to be contaminated. As before, we distribute a large number of particles in a large area in the northwestern Pacific on a fixed date and advected them backward in time. In the previous code, the particles, which crossed the yellow rectangular around the FNPP (Fig.1a) in the past for the period from the day of accident, March 11, 2011 to May 18, 2011, have been marked by the yellow color on the corresponding Lagrangian map. The particles, which were present in that area and leaved it after May 18, have not been colored in yellow. However, those particles also have a risk to be highly contaminated and should be specified as yellow ones. The present code specifies all those particles as yellow ones. As the result, some "white" waters, which have not been specified previously, now have been specified to came from the yellow area around the FNPP with a high risk to be contaminated.

We've cardinally rewritten Secs. 3.1, 3.2 and 3.3 to compare more clearly our simulation results with the measurements by Buesseler2012, Kaeriyama2013, Kuramoto2014 and Budyansky2015. With this aim we imposed on Figs.2c, 2d, 4a and 4d locations of stations with measured values of the cesium concentration levels in collected surface seawater samples in 2011 and 2012.

When working on a revised version of our ms, we have found the paper by Kumamoto, Y. et al. Southward spreading of the Fukushima-derived radiocesium across the Kuroshio Extension in the North Pacific. Sci. Rep. 4, 4276; DOI:10.1038/srep04276 (2014). Seawater samples for radiocesium measurements in the frontal area have been collected during the R/V 'Mirai' cruise in the very beginning of February 2012. We used this new possibility to compare our simulations with this new data. We imposed on the simulated Lagrangian map in Fig.4a locations of stations to the north of the Kuroshio Extention (>36N) with measured levels of the cesium concentrations and found a good qualitative correspondence of those measurements with our simulation results 10 months after the accident in the sense that stations with measured background level are in the area of Oyashio ``blue" waters with low risk to be contaminated, whereas stations with comparatively high level of radiocesium concentrations are in the area of Fukushima-derived ``yellow" waters with increased risk of contamination.

Reviewer #2: Recommendation -

Overall I found the paper to be clear and the figures of good quality. Although being too descriptive, the results presented provide good support for the interpretation of the in-situ observations and could be eventually worth publication. However, I do not recommend the paper to be published in its current state.

Responses to the Second Reviewer's report (Major comments)

1. Cited from the referee's report

As it is, a large portion of the paper is dedicated to the description of the results from previous ocean campaigns which have been already described in previous publications (Buessler et al. 2012; Prants et al. 2014; Kaeriama et al. 2013). The originality of the manuscript needs to be improved. For instance, the authors should provide more details on both a) the computation of Lagrangian diagnostics; b) the eddy identification and tracking.

Our response

We removed from the revised test in Sec.3.1. all the paragraph describing our previous results (Prants et al. 2014). We included in the text a short description of in situ measurements of concentration of Fukushima-derived radiocesium in 2011 and 2012 and impose locations of corresponding stations in the cruises by Buessler, Kaeriama and Budyansky for ease of comparison of observations with our simulation. The computation of Lagrangian diagnostics is described in the second part of Sec.2 Data and methodology on the whole page 3. We have added there the following text describing the eddy identification and tracking as the reviewer asked:

'The altimetry-based Lagrangian maps allow accurately identify and track mesoscale eddies and document their transformation due to interactions with currents and other eddies. Inspecting daily-computed Lagrangian maps for a long period of time (for two years in this paper) and computing stagnation elliptic points daily, one can track the fate of any eddy if it is sufficiently large and long lived (more than a week). The Lagrangian diagnostics is more appropriate with that aim than the commonly used techniques because the Lagrangian maps are imprints of history of water masses involved in the vortex motion whereas vorticity, the Okubo-Weiss parameter and similar indicators are ``instantaneous'' snapshots (see Ref.~\citep{Prants2016} for comparison).'

2. Cited from the referee's report

Regarding the Lagrangian diagnostics: little is said other than that they are derived from AVISO velocity fields. Are the trajectories derived using time varying fields? Are the velocity fields interpolated in both sapce and time? If so, how?

Our response

We've edited the text in the beginning of Sec.2 Data and methodology as follows:

'All the simulation results are based on integrating equations of motion for a large number of synthetic particles (tracers) advected by the AVISO velocity field $\frac{d\lambda}{dt} = u(\lambda, \varphi, t), \frac{d\varphi}{dt} = v(\lambda, \varphi, t)$, where \mathcal{U} and \mathcal{V} are angular zonal and meridional velocities, φ and λ are latitude and longitude, respectively. The altimetry-based velocities were obtained from the AVISO database ($|url\{aviso.altimetry.fr\}$) archived daily on a $1/4^{\circ} \times 1/4^{\circ}$ grid. The velocity field has been interpolated using a bicubical spatial interpolation and third order Lagrangian polynomials in time. In integrating Eqs.~\ref{adveq} we used a fourth-order Runge-Kutta scheme with an integration step of 0.001 day.'

3. Cited from the referee's report

At which spatial resolution are the Lagrangian particles used to generate the maps in Figures 2 to 4 deployed?

Our response

To generate the maps in Figures 2 to 4 we used 700x700 Lagrangian particles. It is mentioned in the revised text.

4. Cited from the referee's report

Which type of AVISO product was used (dt or nrt; two or all sat merged)?

Our response

The AVISO product 'all sat merged delayed time' was used.

5. Cited from the referee's report

On lines 95 to 107 the authors list a series of what they call "Lagrangian indicators", however, they are never shown in the following section. My suggestion is to include only the ones that are discussed in the rest of the manuscript.

Our response

The manuscript is intended for a wide audience, not only to experts in Lagrangian diagnostics. We prefer to save a single sentence listing different Lagrangian indicators because they could be used to provide additional Lagrangian information on transport and mixing of passive particles and tracers.

6. Cited from the referee's report

Among those the authors briefly mention the Finite-time Lyapunov exponent (FTLE). Again, if such diagnostic is used for any of the analyses described in the manuscript (i.e. for definign the elliptic and hyperbolic points introduced in section 2?), then more details on the background of the FTLE and the way they are defined and computed should be provided.

Our response

We did not apply the FTLE technique in this manuscript and removed mention of the FTLE in the revised text.

7. Cited from the referee's report

More details should also be provided on how the elliptic and hyperbolic points are identified. In the manuscript (lines 163-165) they are simply defined as points of zero velocity. How is the circular and diverging/converging motion around those points identified?

Our response

We have added the following text to clarify that:

'The elliptic points are called stable and the hyperbolic ones are unstable. Their local stability properties are characterized by a standard method by eigenvalues of the Jacobian matrix of the velocity field.'

The circular and diverging/converging motion around those points are also defined by eigenvalues of the Jacobian matrix.

8. Cited from the referee's report

Regarding eddy detection and tracking, even less is provided. If they have been identified from previous studies, it should be explicitly said. Otherwise, more details on the methodology should be given.

Our response

We have added there a short text describing the eddy identification and tracking as the reviewer asked.

'The altimetry-based Lagrangian maps allow accurately identify and track mesoscale eddies and document their transformation due to interactions with currents and other eddies. Inspecting daily-computed Lagrangian maps for a long period of time (for two years in this paper) and computing stagnation elliptic points daily, one can track the fate of any eddy if it is sufficiently large and long lived (more than a week). The Lagrangian diagnostics is more appropriate with that aim than the commonly used techniques because the Lagrangian maps are imprints of history of water masses involved in the vortex motion whereas vorticity, the Okubo-Weiss parameter and similar indicators are ``instantaneous'' snapshots (see Ref.~\citep{Prants2016} for comparison).'

9. Cited from the referee's report

A second major issue is with the interpretation of the results from Figures 2 to 4. If I interpret them correctly, each point in the plot has been advected backward in time for two years and then color coded according to its region of origin. The regions are defined in figure 1a. White points are the ones that do not originate from any of those regions. I do agree with the authors, that such backtracking is quite powerful for understanding the origin of waters trapped within the 3 mesoscale eddies investigated.

However, a 2-year backtracking means also that a portion of the "yellow" waters in the figures could have originated from the FNPP area from dates earlier than the accident, and thus not being necessarily contaminated with radioactive material. Wouldn't a Lagrangian experiment forward in time from the region of the FNPP after the time of the accident provide more suited trajectories/diagnostics to infer the fate of the radioactive material released?

Our response

Cited from the manuscript (p.3, lines 187-190): 'In what follows we specify on the maps "yellow waters" as those which have a large risk to be contaminated because they came from the area just around the FNPP enclosed by the yellow lines in Fig. 1a for the period from the day of the accident, March 11, 2011, to May 18, 2011 when direct releases of radioactive isotopes to the ocean and atmosphere stopped'. So, we did not track waters from dates earlier than the accident.

10. Cited from the referee's report

Specific of Figure 2a, at 144 E between 36 and 38 N, there is a very sharp zonal boundary between the different water masses. To me it appears similar to an artefact (either from data processing or visualization). Can the authors comments/correct that?

Our response

We have modified our simulation code and recalculated all the results with this new code. In updated Fig. 2a the straight zonal boundary along 36.5 N and meridional boundary along 144 E, separating water masses of different origin, are just fragments of the yellow straight lines in Fig.1a restricting a potentially radioactive area around the FNPP. The map in Fig. 2a corresponds to March 26, 2011, only 15 days after the accident. The yellow color marks the tracers that have been inside that area or leaved it for these 15 days. The waters of the other colors near those straight zonal and meridional boundaries moved inside the area. So, these boundaries separate the 'yellow' tracers which were present within the area from those ones which have not yet managed to penetrate inside the area for the 15 days.

We briefly clarified that in the revised version to avoid misunderstanding.

11. Cited from the referee's report

A final issue regards the methodological approach: in several occasion throughout the manuscript the authors refer to their Lagrangian approach as "special" (e.g. line 433). Although I agree with the authors when they claim that this type of diagnostics provide a more condensed, easier to read/interpret information, compared to spaghetti plots of particle trajectories, the approach they propose is not entirely novel, as several studies in recent years (for instance d'Ovidio et al. 2015, Biogeoscience to cite one of the most recent ones) have been based on the analysis of similar Lagrangian diagnostics. I suggest to rephrase some of the sentences describing the approach to make this clearer.

Our response

By the word "special" we mean not 'novel' or 'new' but just a thing 'of a particular or certain sort' (cited from the Oxford Dictionary). For example, a special train is an extra train for special purposes.

12. Cited from the referee's report

Furthermore, it is not clear to me how the presented approach would improve the limitations and uncertainties of deriving Lagrangian trajectories in a chaotic environment (lines 71 to 74). In the paragraphs from lines 75 to 85, the authors seem to hint to that the adopted Lagrangian approach is more robust because it does not require "a precise solution of the Navier-Stokes equation".

However, any Lagrangian diagnostic is based on trajectories which require a velocity field to be derived. Later in the manuscript, the author further remark the robustness of their approach stating that it is based on a "statistically significant number of particles". However, other than a large number of particles, shouldn't a statistical Lagrangian approach include also a random-walk term of some sort to simulate sub-grid diffusive-like processes? In my opinion, increasing the number of advected particles, while maintaining the same velocity field and the same equations as a coarser experiment, will provide more detailed results, but not significantly different than a coarser experiment (see for instance Hernandez-Carrasco et al., 2011, Ocean Modelling). These two aspects should be clarified by the authors.

Our response

In this manuscript we don't claim that 'the Lagrangian approach is more robust than a precise solution of the Navier-Stokes equation' (cited from the referee's report). We only claim along with many other authors cited that the Lagrangian approach allows to find more or less robust material structures in chaotic flows without a precise solution of the Navier-Stokes equations (see the first paragraph on p.2, the right column).

We agree, in principle, with the referee that 'increasing the number of advected particles, while maintaining the same velocity field and the same equations as a coarser experiment, will provide more detailed results, but not significantly different than a coarser experiment' (cited from the referee's report). However, obtaining 'more detailed results' is crucial for a specific problem we consider in this ms, tracking of Fukushima-contaminated waters and comparing the simulation results with observation ones.

As to introducing 'a random-walk term of some sort to simulate sub-grid diffusive-like processes' (cited from the referee's report). For chaotic systems, the introduction of a random-walk term in the equations of motion seems to make little sense, because Lyapunov instability of trajectories quickly amplifies noise of any computation method (which is always present), and of a finite accuracy of the representation of real numbers.

Minor comments

13. Cited from the referee's report

The term "Lagrangian particles" should replace "tracers" in several instances in the manuscript.

Our response

We remained the term "tracers" unchanged when speaking about 'colored' particles originated from specified places and replaced it by "Lagrangian particles" in the other cases.

14. Cited from the referee's report

Line 66: if possible I would add some references to the Lagrangian studies which focused on the Horizon accident in the Gulf of Mexico. I am sure that not all of them were only based on the analysis of "spaghetti-like" plots.

Our response

This and the next paragraphs have been slightly edited for more clarity and the following references have been added:

Mezic2010: Mézic IS, Loire VAF, Hogan P (2010) A new mixing diagnostic and the Gulf of Mexico oil spill. Science 330:489.

Olascoaga2012: M. J. Olascoaga and G. Haller}, Forecasting sudden changes in environmental pollution patterns, Proc. National Academy of Sciences, 2012, v.109, p.4738.

Huntley2011: Huntley HS, Lipphardt B, Kirwan A (2011) Surface drift predictions of the deepwater horizon spill: The Lagrangian perspective. Geophys Monogr Ser 195:179–195.

The text now reads as follows:

'The standard approach in simulating transport phenomena like propagation of oil after the explosion at the Blue Horizon mobile drilling rig in the Gulf of Mexico in April 2010 and propagation of radioactive isotopes after the accident at the FNPP is to run global or regional numerical models of circulation to simulate propagation of pollutants and try to forecast their

trajectories. The outcomes provide ``spaghetti-like'' plots of individual trajectories which are hard to interpret. Moreover, majority of trajectories in a chaotic environment are very sensitive to small and inevitable variations in initial conditions. Those trajectories are practically unpredictable even over a comparatively short time.

The specific Lagrangian approach, based on dynamical systems theory, has been developed in the last decades with the aim to find more or less robust material structures in chaotic flows governing mixing and transport of Lagrangian particles and creating transport barriers of a contaminant across them preventing propagation \citep[for reviews see][] {Samelson, MS06, KP06, Haller ARFM2015}. Identification of such structures in the ocean would help to predict for a short and medium time where a contaminant will move even without a precise solution of the Navier\mdash Stokes equations. This approach has been successfully used simulating propagation in Gulf in of oil the Mexico of \citep{Mezic2010,Huntley2011,Olascoaga2012} and propagation of Fukushima-derived radionuclides in the Pacific ocean \citep{DAN11,DSR201,Prants2014}.'

15. Cited from the referee's report

Fig 1a: I would plot SST rather than velocity magnitude (since temperature variation and fronts are repeatedly used in the introduction), and I would remove the indication of elliptic and hyperbolic points since in a climatological velocity field they do not represent mesoscale structures. **Our response**

Figure 1a with the averaged AVISO velocity field from 1993 to 2016 gives an image of persistent mesoscale features in the study area governing the large-scale transport and mixing. SST does not provide that. We removed the indication of elliptic and hyperbolic points from that figure.

16. Cited from the referee's report

Line 79: Diffusion will always occur. It is the mixing induced by advection that is reduced across transport barriers.

Our response

We mean there a diffusive-like propagation, not a molecular diffusion. However, in order to avoid a misunderstanding we removed the term 'diffusive-like' from the text and edited the sentence as:

'The specific Lagrangian approach, based on dynamical systems theory, has been developed in the last decades with the aim to find more or less robust material structures in chaotic flows governing mixing and transport of Lagrangian particles and creating transport barriers preventing propagation of a contaminant across them.'

17. Cited from the referee's report

Line 123: replace "twofold" with "threefold" since the same paragraph contain a "Firstly...", a "Secondly..." and a "Finally..."

Our response

Done.

18. Cited from the referee's report

Line 147-149: AVISO provide the geostrophic component of the real near-surface velocities. **Our response**

Done.

19. Cited from the referee's report

Line 165-167: The sentence should be moved after line 169, since it refers to elliptic points only.

Our response

Done.

20. Cited from the referee's report

Line 176: Several studies in the last few years have shown indeed that LCS and hyperbolic points can be identified and tracked for several days from in-situ observations: Haza et al. 2010, Ocean Dynamics; Nencioli et al 2011, JGR-Oceans; Olascoaga et al. 2013, GRL. They should be cited here.

Our response

The following sentence has been added just after Lone 176:

'The hyperbolic points and their attracting and repelling manifolds have been recently identified with the help of drifter's tracks in the Gulf of La Spezia in the northwestern Mediterranean Sea \citep{Haza2010}, in the Gulf of Mexico \citep{Olascoaga2013} and in the northwestern Pacific \citep{Prants2016}.'

Haza2010: Haza, A.C., Özgökmen, T.M., Griffa, A. et al. Transport properties in small-scale coastal flows: relative dispersion from VHF radar measurements in the Gulf of La Spezia. Ocean Dynamics (2010) 60: 861. doi:10.1007/s10236-010-0301-7

Olascoaga2013: M. J. Olascoaga et al. Drifter motion in the Gulf of Mexico constrained by altimetric Lagrangian coherent structures. GEOPHYSICAL RESEARCH LETTERS, V. 40, doi:10.1002/2013GL058624, 2013.

Prants2016: S.V. Prants, V.B. Lobanov, M.V. Budyansky, M.Yu. Uleysky. Lagrangian analysis of formation, structure, evolution and splitting of anticyclonic Kuril eddies. Deep Sea Research I. V.109 pp.61–75 (2016). DOI: 10.1016/j.dsr.2016.01.003

21. Cited from the referee's report

Line 381: were the ARGO floats regular float, or were characterized by specific configurations?

Our response

They are regular floats. We corrected the text

"by tracks of surface drifters and diving Argo floats available at the sites www.argo.net and aoml.noaa.gov/phod/dac, respectively"

to be

"by tracks of surface drifters and diving Argo floats available at the sites aoml.noaa.gov/phod/dac and www.argo.net, respectively."

Lagrangian simulation and tracking of the mesoscale eddies contaminated by Fukushima-derived radionuclides

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Abstract. A Lagrangian methodology is elaborated to simulate, track, document and analyze origin and history of water masses in ocean mesoscale features. It is aimed to distinguish water masses inside the mesoscale eddies originated

- ⁵ from the main currents in the Kuroshio Oyashio confluence zone. Computing trajectories for a large number of synthetic tracers Lagrangian particles advected by the AVISO velocity field after the Fukushima accident, we identify and track the mesoscale eddies which have been sampled in the cruises in the cruises
- ¹⁰ 2011 and 2012 and estimate their risk to be contaminated by Fukushima-derived radionuclides. The simulated results are compared with *in situ* measurements to be in a good qualitative correspondence.

1 Introduction

- ¹⁵ High tsunami waves after the Tohoku earthquake on March 11, 2011 damaged the cooling system of the Fukushima Nuclear Power Plant (FNPP). Due to lack of electricity, it was not possible to cool nuclear reactors and the fuel storage pools that caused numerous explosions at the FNPP (for ²⁰ details see Povinec et al., 2013). The Fukushima accident was classified at the maximum level of 7, similar to the Chernobyl accident which happened in 1986 in the former
- Soviet Union. Radionuclides were released from the FNPP through two major pathways, direct discharges of radioac-²⁵ tive water and atmospheric deposition onto the North Pacific
- Ocean. Indirect estimation of that deposition is in the range 6.4–35 PBq (Kumamoto et al., 2014). The total amount of 137 Cs isotope released into the ocean was estimated to be $3.6 \pm 0.73.6 \pm 0.7$ PBq by the end of May 2011 (Tsumune ³⁰ et al., 2013).

A few special research vessel (R/V) cruises have been conducted just after the accident and later to measure radioactivity in sea water, zooplankton, fish and in other marine organisms. ¹³⁷Cs and ¹³⁴Cs isotopes with 30.17 yr and 2.06 yr half-life, respectively, have been detected over ³⁵ a broad area in the western North Pacific in 2011 and 2012 (Honda et al., 2012; Buesseler et al., 2012; Inoue et al., 2012a, b; Tsumu ¹³⁷Cs concentration levels off Japan before the accident were estimated at the background level to be 1–2 Bq m⁻³ or $\simeq 0.001-0.002$ Bq kg⁻¹ mBq/kg, while ¹³⁴Cs was not detectable. Because of a comparatively short half-life time, any measured concentrations of ¹³⁴Cs could only be Fukushima derived.

The studied area is shown in Fig. 1a. It is known as a Kuroshio – Ovashio confluence zone or a subarctic frontal 45 area (Kawai, 1972). The Kuroshio Extension prolongs the Kuroshio Current which turns to the east at about 35° N and flows as a strong meandering jet constituting a front separating the warm subtropical and cold subarctic waters. It is a region with one of the most intense air-sea heat exchange and 50 the highest eddy kinetic energy level. The Kuroshio-Oyashio Kuroshio-Oyashio confluence zone is populated with a plenty of mesoscale eddies that transfer heat, salt, nutrients, carbon, pollutants and other tracers across the ocean. They originate, besides from the Kuroshio Extension, from the 55 Tsugaru Warm Current, flowing between the Honshu and Hokkaido islands, and from the cold Oyashio Current flowing out of the Arctic along the Kamchatka Peninsula and the Kuril Islands (Fig. 1a). The lifetime of those eddies ranges from a few weeks to a few years. 60

Lagrangian tools have been successfully used

The standard approach in simulating transport phenomena including like propagation of oil after the explosion at the



Figure 1. a) The AVISO velocity field in the Kuroshio–Oyashio confluence zone averaged from 1993 to 2016. TsS stands for the Tsugaru Strait. Elliptic and hyperbolic stagnation points with zero mean velocity are indicated by triangles and crosses, respectively, with up- and downward oriented triangles denoting anticyclones and cyclones, respectively. Location of the FNPP is shown by the radioactivity sign. The area Area just around the FNPP is shown by the yellow lines. b) The velocity field on August 24, 2011 with the Tohoku (TE) and Hokkaido (HE) eddies studied in the paper and with tracks of some available drifters (the red circles) and ARGO floats (the green stars) to be present in the area at that time. Elliptic and hyperbolic stagnation points with zero mean velocity are indicated by triangles and crosses, respectively, with up- and downward oriented triangles denoting anticyclones and cyclones, respectively.

Blue Horizon mobile drilling rig in the Gulf of Mexico in April 2010 and propagation of radioactive isotopes after the accident at the FNPP . The standard approach is to run global or regional numerical models of circulation to simulate prop-⁵ agation of pollutants and try to forecast their trajectories. The outcomes provide "spaghetti-like" plots of individual trajectories which are hard to interpret. Moreover, majority of trajectories in a chaotic environment are very sensitive to small and inevitable variations in initial conditions. Those trajectories are practically unpredictable even over a comparatively ¹⁰ short time.

The specific Lagrangian approach, based on dynamical systems theory, has been developed in the last decades with the aim to find more or less robust material structures in chaotic flows governing mixing and transport of tracers 15 Lagrangian particles and creating transport barriers preventing diffusive-like propagation of a contaminant across them (for reviews see Samelson and Wiggins, 2006; Mancho et al., 2006; Koshel' and Prants, 2006; Haller, 2015). Identification of such structures in the ocean would help to 20 predict for a short and medium time where a contaminant will move even without a precise solution of the Navier-Stokes equations. This approach has been successfully used in simulating propagation of oil in the Gulf of Mexico (Mezić et al., 2010; Huntley et al., 2011; Olascoaga and Hallers 2012) and propagation of Fukushima-derived radionuclides Pacific in the ocean (Prants et al., 2011b; Budyansky et al., 2015; Prants et al., 2014).

The present authors have developed the appropriate La- 30 grangian tools for tracking origin, history and fate of water masses advected by analytic, altimetric and numerical velocity fields generated by eddy-resolved regional circulation models (Budyansky et al., 2009; Prants et al., 2011b, a; Prants, 2013; Prants et al., 2013; Prants, 2014; Budyansky 35 et al., 2015). Each elementary volume of water can be attributed to physico-chemical properties (temperature, salinity, density, radioactivity, etc.) which characterize this volume as it moves. In addition, each water parcel can be attributed to more specific characteristics as trajectory's func- 40 tions that carry key data but are not physico-chemical properties. We call them "Lagrangian indicators". They are, for example, distance passed by a fluid particle for some period of time; absolute, zonal, and meridional displacements of particles from their original positions; the number of 45 their cyclonic and anticyclonic rotations; time of residence of fluid particles inside a given area and the exit time out off that area; the number of times particles visited different places in a studied region. The commonly used finite-time Lyapunov exponent is one of the Lagrangian indicators in 50 our terminology.

The Lagrangian indicators contain information about the origin, history and fate of the corresponding water masses and allow to identify water masses moving coherently, either propagating together or rotating together. Even if adjacent waters are indistinguishable, say, by temperature, and the satellite SST images indicate no thermal front, the corresponding water masses can be distinguishable, for example, by their origin, travelling history and other factors. The Lagrangian indicators are computed by integrating advection 60 equations (1) for a large number of synthetic tracers-particles

forward and backward in time. When integrating (1) forward in time one computes particle's trajectories to know the fate of the corresponding tracers particles and when integrating (1) backward in time one could know where the tracers s particles came from and the history of their travel.

The purpose of this paper is **twofold**. Firstly, we develop a Lagrangian methodology in order to track and document the origin and history of water masses constituting prominent mesoscale feature. It allows to distinguish ¹⁰ water masses inside mesoscale eddies originated from the main currents in the Kuroshio–Oyashio confluence zone. Secondly, we apply that methodology in order to identify and track the mesoscale eddies, advected by the altimetric AVISO velocity field, with a risk to be contaminated ¹⁵ by Fukushima-derived radionuclides. Finally, the simulation

results are compared qualitatively with *in situ* sampling of those eddies in the R/V cruises. The location and form of the simulated eddies are verified, when possible, by tracks of surface drifters and diving <u>Argo ARGO</u> floats available

²⁰ at the sites and aoml.noaa.gov/phod/dac and www.argo.net, respectively.

2 Data and methodology

Geostrophic velocities were obtained from the AVISO database () archived daily on a $1/4^\circ\times 1/4^\circ$ grid. We

²⁵ integrate equations All the simulation results are based on integrating equations of motion for a large number of synthetic particles (tracers) advected by the interpolated AVISO velocity field

$$\frac{d\lambda}{dt} = u(\lambda, \varphi, t), \qquad \frac{d\varphi}{dt} = v(\lambda, \varphi, t), \tag{1}$$

- ³⁰ where *u* and *v* are angular zonal and meridional velocities, φ and λ are latitude and longitude, respectively. The altimetry-based velocities were obtained from the AVISO database (aviso.altimetry.fr) archived daily on a $1/4^{\circ} \times 1/4^{\circ}$ grid. The velocity field has been interpolated using a ³⁵ bicubical spatial interpolation and third order Lagrangian
- polynomials in time. In integrating Eqs. 1 we used a fourth-order Runge – Kutta scheme with an integration step of 0.001 day. Altimetry data provide the velocity field which is a geostrophical approximation to component of the real
- ⁴⁰ near-surface velocities. The results of our altimetry-based Lagrangian statistical analysis are expected to be valid on a mesoscale where the AVISO velocity field may be considered to be a good approximation.

In order to display the enormous amount of information, ⁴⁵ we compute Lagrangian maps which are plots of a specific Lagrangian indicator versus particle's initial positions. The region under study is seeded with a large number of tracers Lagrangian particles whose trajectories are computed for a given period of time. The results obtained are processed to

50 get a data file with the field of a specific Lagrangian indi-

cator in this area. Finally, its values are coded by color and represented as a map in geographic coordinates.

It is informative to impose "instantaneous" stagnation elliptic and hyperbolic points on the Lagrangian maps. We mark them by triangles and crosses, respectively. They are 55 points with zero velocity which are computed daily with the AVISO velocity field. Up(down)ward orientation of one of the triangle's top means anticyclonic (cyclonic) rotations of water around them The elliptic points are called stable and the hyperbolic ones are unstable. Their local stability 60 properties are characterized by a standard method calculating eigenvalues of the Jacobian matrix of the velocity field. The elliptic points, situated mainly in the centers of eddies, are those points around which the motion is stable and circular. Up(down)ward orientation of one of the 65 triangle's top on the maps means anticyclonic (cyclonic) rotations of water around them. The hyperbolic points, situated mainly between and around eddies, are unstable with the direction have stable manifolds along which water parcels converge to such a point and another direction 70 unstable manifolds along which they diverge. The stagnation points are moving Eulerian features and may undergo bifurcations in the course of time. In spite of nonstationarity of the velocity field, some of them may exist for weeks and much more. The hyperbolic points and their attracting 75 and repelling manifolds have been recently identified with the help of drifter's tracks in the Gulf of La Spezia in the northwestern Mediterranean Sea (Haza et al., 2010), in the Gulf of Mexico (Olascoaga et al., 2013) and in the northwestern Pacific (Prants et al., 2016). 80

The altimetry-based Lagrangian maps allow accurately identify and track mesoscale eddies and document their transformation due to interactions with currents and other eddies. Inspecting daily-computed Lagrangian maps for a long period of time (up to two years in this paper) and computing stagnation elliptic points daily, one can track the fate of any eddy if it is sufficiently large and long lived (more than a week). The Lagrangian diagnostics is more appropriate with that aim than the commonly used techniques because the Lagrangian maps are imprints of history of water masses involved in the vortex motion whereas vorticity, the Okubo–Weiss parameter and similar indicators are "instantaneous" snapshots (see Olascoaga et al., 2013; Prants et al., 2016, for comparison).

Being motivated by the problem of identification of Fukushima-contaminated waters in the core and at the periphery of persistent mesoscale eddies in the area, we develop in this paper a special Lagrangian technique oriented to distinguish water masses of a different origin inside the eddies with a risk to be contaminated. With this aim we specify, besides Fukushima-derived waters, water masses originated from the main currents in the Kuroshio – Oyashio confluence zone. The integration was has been performed backward in timefor two years starting from a date indicated on 105 the corresponding simulated Lagrangian map. . We removed from consideration all the particles entered into any AVISO grid cell with two or more corners touching the land because of inaccuracy of the altimetry-based velocity field there and 5 in order to avoid artifacts.

In what follows we specify on the maps "yellowwaters" "waters as those which have a large risk to be contaminated because they came after the accident from the area just around the FNPP enclosed by the yellow straight lines

- ¹⁰ in Fig. 1afor the period from the day of the accident, March 11, 2011, to May 18, 2011 when direct releases of radioactive isotopes to the ocean and atmosphere stopped... The "redwaters" <u>waters</u> are salty and warm Kuroshio waters. To be more exact, they came from the red zonal line
- ¹⁵ (34.5°N, 139°E–144°E) in Fig. 1a crossing the Kuroshio main jet. The "blackwaters" "waters came from the warm Tsushima Current flowing via the Tsugaru Strait out off the Japan Sea and crossed that strait (the black line with $40^{\circ}N-43^{\circ}N$, 141.55°E). The "bluewaters" "waters are
- ²⁰ more fresh and cold waters originated from the Oyashio Current and crossed the blue zonal line (48° N, 153° E-159° E) shown in Fig. 1a. The "white" waters on the Lagrangian maps have not been specified to be originated from one of the segments mentioned above. They could reach their places on ²⁵ the maps from anywhere besides those segments.

²⁵ the maps from anywhere desides those segments

We are interested in advective transport for a comparatively long period of time, up to two years. It is hardly possible to simulate adequately motion of a specified passive particle in a chaotic flow, but it is possible to reproduce trans-

- ³⁰ port of statistically significant number of particles. Our results are based not on simulation of individual trajectories but on statistics for hundreds of thousands of tracers490,000 Lagrangian particles. We cannot, of course, guarantee that we compute "true" trajectories for individual tracersparticles.
- ³⁵ The description of general pattern of transport for many thousands of tracers half a million particles is much more robust. However, we do not try to simulate quantitatively concentration of radionuclides or estimate the content of water masses of different origin inside the studied eddies.

40 3 Results

A number of mesoscale eddies was few mesoscale eddies were present in the studied area to the day of the accident. The cyclonic eddies with the centers at downward-oriented triangles on the Lagrangian maps prevailed in the area to the so north of the Subarctic Front, the boundary between the subarctic ("blue") and subtropical ("red") waters in Fig. 2. The anticyclonic eddies with the centers at upward-oriented triangles prevailed to the south of the front.

The large anticyclonic Tohoku eddy (TE) with the center ⁵⁰ at around 39°N, 144°E in March 2011 was has been sampled after the accident in the two R/V cruises in June (Buesseler et al., 2012) and July 2011 (Kaeriyama et al., 2013) to have large concentrations of ¹³⁷Cs , and ¹³⁴Cs. The anticyclonic Hokkaido eddy (HE), genetically connected with the TE, was born in the middle of May 2011 with the center at around 40° N, 145° Eand captured some Fukushima. After that it captured some contaminated water from the TE. It was sampled partly has been sampled in the end of July 2011 (Kaeriyama et al., 2013).

The anticyclonic Tsugaru eddy (TsE) was genetically connected with the HE. It was born in the beginning of February 2012 with the center at around 41.9° N, 148° E and captured some Fukushima contaminated water from the HE. The TsE was has been sampled in the R/V "*Professor Gagarinskiy*" cruise on July 5, 2012 to have higher concentrations of ¹³⁷Cs and ¹³⁴Cs over the background level at the surface and in intermediate depths (Budyansky et al., 2015). All these eddies will be studied in this section from the Lagrangian point of view in order to simulate and document track by which transport pathways they could gain water masses from the Fukushima area and water masses of other origin and to compare qualitatively the simulation results with the *in situ* measured onesmeasurements.

3.1 The Tohoku eddy

We were able to track with daily computed tracked with 75 daily-computed Lagrangian maps the birth, metamorphoses and decay of the mesoscale anticyclonic TE. It was born in the middle of May 2010 with the elliptic point at around 38° N, 144° E at that time as the result of interaction between of a warm anticyclonic Kuroshio ring and with a cyclone with 80 mixed Kuroshio and Oyashio core waters. It has interacted with another eddies almost for a year with multiple splittings splitting and merging in the area to the east off the Honshu Island. Just after the accident, it begun to gain "yellowwater" " water from the area around the FNPP with a high risk of 85 contamination. That eddy is clearly seen in earlier simulation just after the accident in Fig. 3b by Prants et al. (2011b) and on the Lagrangian map in Fig. 2a as a red patch labeled as TE with the center at 39° N, 144° E on March 26, 2011.

The maps in Fig. 2 and in the subsequent figures have been 90 computed as it was explained in Sec. 2. The red color in the core of the TE means that its core water was of a Kuroshio subtropical origin. More exactly, the red tracers came for two years in the past to their places on the map from the red line segment in Fig. 1a crossing the Kuroshio jet. In March 95 2011 "yellowwater" " water, coming from the area around the FNPP with a comparatively high risk to be contaminated, wrapped round the TE. A thin streamer of Tsugaru "blackwater" "water, coming from the black line segment in Fig. 1a, wrapped a periphery of the TE to the end of March. 100 "Yellowwaters" " waters propagated gradually to the east and south due to a current system system of currents sometimes wrapping round the eddies to be present in the area. The straight zonal boundary along 36.5°N and meridional boundary along 144°E, separating water masses of different 105



Figure 2. The Lagrangian maps show evolution of the Tohoku eddy (TE) after the FNPP-accident to the days of its sampling and the origin of waters in its core and at the periphery. The red, black and blue colors specify waters the tracers which came for two years in the past to their places on the maps from the Kuroshio, Oyashio and Tsushima currents, respectively, more exactly, from the corresponding line segments shown in Fig. 1a. The yellow color marks the Lagrangian particles coming from the area around the FNPP in Fig. 1a after the day of the accident on March 11, 2011. The TE has been sampled on June 10 and 11, 2011 (Buesseler et al., 2012) by Buesseler et al. (2012) along the colored transect at 35.5° N $- 38^{\circ}$ N, 144° E shown in panel c) and in the end of July 2011 (Kaeriyama et al., 2013) by Kaeriyama et al. (2013) along the colored transect 35° N $- 41^{\circ}$ N, 144° E shown in panel d). The locations of stations with collected by Buesseler et al. (2012) and (Kaeriyama et al., 2013) surface seawater samples with measured radiocesium concentrations at the background level are indicated by the green diamonds. Stations, where the concentrations have been measured to be much higher, are marked by the magenta diamonds.

origin in Fig. 1a on March 26, 2011, are just fragments of the yellow straight lines in Fig. 2a restricting the area around the

FNPP. These boundaries separate the "yellow" tracers which were present within the area from those ones which have not



Figure 3. a) and b) The Lagrangian maps show evolution of the Hokkaido eddy (HE) after the FNPP accident to the days of its sampling and the origin of waters in its core and at the periphery. c) and d) A fragment of the track of the drifter no. 39123 is indicated by the full circles for two days before the day indicated with the size of circles increasing in time. Tracks of the three ARGO floats are shown by the stars. The largest star corresponds to the day indicated and the other ones show float 's location positions' each 5 days before and after that date.

yet managed to penetrate inside the area for 15 days after the accident.

In April and May 2011 the TE had a sandwich-like structure with the red Kuroshio subtropical core belted with

⁵ a narrow streamer of Fukushima "yellowwaters"" waters which, in turn, was encircled by a red streamer of Kuroshio subtropical water (Fig. 2b). A new eddy configuration appeared to the end of May in Fig. 2b with the TE interacting with a eyelone (the elliptic point "blue" cyclone with the contact at 20.0° N 144.7° E) and a newhore "vallew" entire.

¹⁰ center at 39.9° N, 144.7° E) and a newborn <u>"yellow</u>" anticyclone which we call the Hokkaido eddy (HE) with the center at 40.4°N, 145.5°E. The core of the that cyclone consisted of a "blue" subarctic Oyashio water with low risk to be contaminated, but the HE core water came from the area around the FNPP with a high risk to be contaminated.

In the course of time the TE moved gradually to the southand was . Its periphery has been sampled in the beginning of June (Buesseler et al., 2012) and by Buesseler et al. (2012), and the whole eddy has been crossed in the end of July 2011 (Kaeriyama et al., 2013) by 20 Kaeriyama et al. (2013). Fukushima-derived cesium isotopes have been measured on June 10 and 11 June during



Figure 4. The Lagrangian maps show evolution of in the Tsugaru eddy (TsE) study area in the first half of 2012 and the origin 2012. a) The locations of waters stations in its core and the beginning of February with collected by Kumamoto et al. (2014) surface seawater samples with measured radiocesium concentrations at the periphery background level (the green diamonds) and with higher concentration levels (the magenta diamonds). That b) – d) The Lagrangian maps show evolution of the Tsugaru eddy (TsE) which was born on February 4, 2012 (panel a) after splitting of the HE and sampled in the R/V "Professor Gagarinskiy" cruise by Budyansky et al. (2015) at station 84 on July 5, 2012 (Budyansky et al., 2015) to have higher increased radiocesium concentrations of and over the background level at (the surface and magenta diamond in intermediate depthspanel d).

the R/V "*Ka'imikai-o-Kanaloa*" cruise (Buesseler et al., 2012) along the meridional transect shown in Fig. 2e where -144° E meridional transect where the cesium concentrations have been found to be in the range from ⁵ the background level, $1.4\pm0.2C_{137} = 1.4-3.6$ mBq/kg

(station stations 13 and 14), to a high level of 173.6 ± 9.9 up to $C_{137} = 173.6 \pm 9.9$ mBq/kg (station 10). The ratio 134 Cs/ 137 Cs was close to 1.

Prants et al. (2014) have simulated backward in time advection of the tracers placed on a material line along 10

that transect. Computing how many times they visited each cell in the studied area from March 11 to April For ease of comparison, we mark in Fig. 2c by the green diamonds the locations of stations 13 and 14 with collected ⁵ surface seawater samples by Buesseler et al. (2012) in which

- the cesium concentrations have been measured to be at the background level. The stations 10, 2011, they plotted so-called tracking maps. Traces of the particles, chosen at the green segment 11 and 12, where the concentrations have
- 10 been found to be much larger, are indicated by the magenta diamonds. Our simulation in Fig. 2c , have been found on the both sides of the Kuroshio Extension jet, whereas traces of particles at the red one have been found on the northern side of the jet only (see Fig. 6 by Prants et al. (2014)). The
- 15 measured concentrations of along the green segment, were at the background level, in shows that stations 13 and 14 on the days of sampling have been located in "red" and "white" waters with a low risk to contain Fukushima-derived radionuclides.
- Transport and mixing at and around stations 10, 11 and 12 20 with high measured values of the range 1.4-3.6 mBqcesium concentrations by Buesseler et al. (2012) have been governed mainly by the interaction of the TE with the "yellow" mesoscale cyclone with the center at 37.2° N,
- ²⁵ 142.8°E. This cyclone formed in the area in April and captured "yellow" waters with a high risk of contamination. Unfortunately, it has not been sampled in the R/kg (Buesseler et al., 2012), because the corresponding particles were advected by the Kuroshio jet. Density of traces of the
- 30 "red" particles has been shown by Prants et al. (2014) to be comparatively high in the area around the FNPP. This finding correlates with the measurements along the red transect where the concentrations of Fukushima-derived radiocesium were in the range 21.9-173.6V "Ka'imikai-o-Kanaloa"
- 35 cruise. The surface seawater samples at stations 10, 11 and 12, have been collected on the days of sampling at the eastern periphery of that cyclone and at the southern periphery of the TE with the "yellow" streamer there. Station 10 with the highest measured level of the ¹³⁷Cs concentration,
- 40 $C_{137} = 173.6 \pm 9.9$ mBq/kg. The red segment crosses only the very periphery of the TE visible, was located at 38° N, 144°E inside the wide streamer of "yellow" water around the TE. Stations 11 and 12 with $C_{137} = 103.7 \pm 5.9$ mBq/kg and $C_{137} = 93.6 \pm 4.9$ mBq/kg, respectively, have been
- 45 located within the narrow streamers with "yellow" simulated water in Fig. 2c as a deformed patch with the elliptic point at, with the intermitted with narrow streamers of "red" core and "yellow water" around. water. So, we estimate the risk to find Fukushima-derived radionuclides there (the magenta
- 50 diamonds) to be much higher than at stations 13 and 14 (the green diamonds) and it is confirmed by a qualitative comparison with measured data.

A specific configuration of mesoscale eddies occurred in the area to the north-northeast of the FNPP to the end of July 55 2011, the days of sampling by Kaeriyama et al. (2013) along the 144° E meridian from 35° N to 41° N in the R/V "Kaiun maru" cruise(Kaeriyama et al., 2013). That transect is shown in Fig. 2d. It crosses the TE and the cyclone with "blue" Oyashio water, which is genetically linked to the evelone shown "blue" cyclone at 39.9° N, 144.7° E in Fig. 2b. The 60 transect also crosses partly the periphery of the anticyclonic HE. The measured ¹³⁷Cs concentrations in surface seawater samples at the stations C43-C55 have been found to be in the range from the background level, 1.9 ± 0.4 mBg/kg, (station C52) to a much higher level of 153 ± 6.8 mBq/kg (station 65 C47). The colored tracking maps in Fig. 5 by Prants et al. (2014), colored as the corresponding segments in Fig. 2d, show where the simulated tracers of that transect were walking from March 11 to April 10, 2011. 2011 being advected by the AVISO velocity field. 70

The risk of radioactive contamination of the orange markers was shown markers placed at 36° N-36.5° N was estimated by Prants et al. (2014) to be small, because they were have been advected mainly by the Kuroshio Current from the southwest to the east (the corresponding concen-75 trations have been measured by Kaeriyama et al. (2013) to be 2-5 mBq/kg(Kaeriyama et al., 2013)). The red segment present simulation in Fig. 2d also shows that stations C51, 52 and 53 (the green diamonds) with the measured cesium concentrations at the background level on the days of 80 sampling by Kaeriyama et al. (2013) have been located in the "red" waters (stations C51 and C53) advected by the main Kuroshio jet from the southwest and in the "white" waters (station C52) between the TE and the jet. Therefore, we estimate a risk to find Fukushima-derived radionuclides there 85 to be comparatively low.

The transect 36.5° N-38° N in Fig. 2d erosses (the red one in Fig. 5 by Prants et al. (2014)) crossed the TE. The ¹³⁷Cs concentrations at the stations C49 and C50 of that segment were transect have been measured to be $36 \pm 3.3_{90}$ and 50 ± 3.6 mBq/kg (Kaeriyama et al., 2013). The highest concentration levels up to 153 ± 6.8 Comparing those results with simulated ones, we note the presence of "yellow" water in the TE core at the locations of those stations. Surface samples at station C48 (38.5°N) have been measured to 95 contain the ¹³⁷Cs concentration to be at the background level 2.7 ± 0.6 mBq/kg (Kaeriyama et al., 2013)have been measured at the stations C46. The corresponding green diamond is located in our simulation in the area with "red" and "white" waters. 100

Inspecting the Lagrangian maps on the days between June 6 and July 28 (not shown), we have found that the "yellow" cyclone with the center at 37.2°N, 142.8°E in Fig. 2c collapsed in the end of June. Its "yellow" core water with a high risk to be contaminated has been 105 wrapped around the neighbor anticyclone TE in the form of a wide yellow streamer visible in Fig. 2d. The highest concentration $C_{137} = 153 \pm 6.8$ mBq/kg has been measured by Kaeriyama et al. (2013) at station C47 (39°N) situated in the area of that streamer. Stations C46 (39.5°N) with 110

 $C_{137} = 83 \pm 5.0 \text{ mBq/kg}$ is situated in the close proximity to a yellow streamer sandwiched between "white" and "black" waters.

- Comparatively high concentration $_5 C_{137} = 65 \pm 4.3$ mBq/kg has been measured by Kaeriyama et al. (2013) at station C45 (40° N) that was on the days of sampling in the core of the "blue" cyclone with the center at 39.7° N, 144.2° E (Fig. 2d). Our simulation shows that it has been formed mainly by Oyashio
- ¹⁰ "blue" waters (with a low risk to be contaminated by Fukushima-derived radionuclides) and partly by "white" waters.

When comparing simulation results in Fig. 2d with the measurements by Kaeriyama et al. (2013), we have found

- ¹⁵ that the simulation consists with samplings at stations C48, 51, 52 and C48 situated at the blue segment and at C4353 in the sense that the cesium concentrations have been measured to be at the background level in those places on the maps where there is no signs of "yellow" water with a high risk
- to contain Fukushima-derived radionuclides. Our simulation consists at least quantitatively also with samplings at stations C47, C44 49 and 50 with high measured levels of the cesium concentrations because the "yellow" water presents there in our simulation.
- However, there is an inconsistency of simulation with samplings at stations C45 on the green segment. The tracking maps in Figs. 5c and d by Prants et al. (2014) show clearly an increased density of traces of the corresponding markers in the area where the maximal leakage of radionuclides
- ³⁰ and C46 where there are practically now yellow tracers but only blue and white ones. The reasons of this inconsistency might be different. In this paper we track only those tracers which were originated from the blue, red and black segments and the yellow rectangular around the FNPP shown in
- ³⁵ Fig. 1a. So we did not specify the origin of white waters. They could reach their places on the maps from anywhere besides those segments and the area around the FNPP. They could in principle contain Fukushima-derived radionuclides deposited at the sea surface from the FNPP directly into the
- 40 ocean and their atmospheric fallout on the ocean surface have been registered from March 11 to April 10, 2011. Thus, the TE has interacted for many months atmosphere after the accident with a number of adjacent eddies and streamers providing transport of contaminated water to the
- ⁴⁵ north, south and east. Its influence to transport and mixing of the contaminated water in the area is evident as well on the tracking maps by Prants et al. (2014) and then being advected by eddies and currents in the area. Moreover, they could be those tracers which have been located inside AVISO grid
- ⁵⁰ cells near the coast around the FNPP just after the accident and then have been advected outside. We removed from consideration all the tracers entered into any AVISO grid cell with two or more corners touching the land because of inaccuracy of the altimetry-based velocity field there and in

55 order to avoid artifacts.

Thus, the white streamers inside the core and at the periphery of the blue cyclone with the center at 39.7° N, 144.2° E nearby stations C45 and C46 with high measured concentrations of cesium by Kaeriyama et al. (2013), could, in principle, contain contaminated water. However, it has not been proved in our simulation by the mentioned reasons.

3.2 The Hokkaido eddy

Now we consider the anticyclonic HE. It was born in the end-middle of May (see the yellow patch in Fig. 2b with the center at 40.3°N, 145.5°E) being genetically linked to the TE. During May, the TE gradually lost a Fukushima "yellowwater" " water from its periphery to form the core of the HE. FigureFig. 3a shows the HE with a yellow core surrounded by modified subtropical "redwater"" water which, in turn, is surrounded by Tsugaru "blackwater"." water

The sampling of that eddy and its periphery by Kaeriyama et al. (2013) along the 144°E meridian (the green segment in Fig. 2d) by Kaeriyama et al. (2013) in the end of July showed a high concentration of both the radiocesium isotopes. comparatively high concentrations, $C_{137} = 60 \pm 4.0$ and 71 ± 4.6 mBq/kg at stations C44 (40.5°N) and C43 (41°N), respectively. Station C43 was located inside the anticyclone HE filled mainly by "yellow" waters, and we estimate the risk to found Fukushima-derived radionuclides there to be large. Station C44 was located at the southern periphery of the anticyclone HE at the boundary between "white" and "blue" waters but in close proximity to a "yellow" streamer.

The location of the HE on August 24, 2011 is shown in the AVISO velocity field in Fig. 1b. To verify the simulated locations of the HE and its form, we plot in Figs. 3c and d fragments of the tracks of a drifter and three ARGO floats captured by that eddy in September 2011.

A fragment of the track of the drifter no. 39123 is shown by the red circles with the size increasing in time for two 90 days before the dates indicated in Figs. 3c and d and decreasing for two days after those dates, i.e. the largest circle corresponds to the drifter 's location position at the indicated date. It was launched after the accident on July 18, 2011 at the point 45.588° N, 151.583° E in the Oyashio Current, ad- 95 vected by the current to the south and eventually captured by the HE moving around clockwise. Fragments of the clockwise tracks of the three ARGO floats are shown in Figs. 3c and d by stars for seven days before and seven days after the indicated dates. The float no. 5902092 was released 100 long before the accident on September 9, 2008 at the point 32.699° N, 145.668° E to the south off the Kuroshio Extension jet and was able to cross the jet and to get far north. The float no. 2901019 was released before the accident on April 19, 2010 at the point 41.723° N, 146.606° E. The float 105 no. 2901048 was released just after the accident on April 10, 2011 at the point 37.469° N, 141.403° E nearby the FNPP.

Our simulation shows that the HE contained <u>after its</u> formation in the middle of May 2011 a large amount of a "yellowwater" with the risk to be" water probably contaminated by the Fukushima-derived radionuclides. This conclu-

 ⁵ sion is supported by an increased concentration of radiocesium measured by Kaeriyama et al. (2013) at its periphery in its core at station C43 by Kaeriyama et al. (2013) in the end of July 2011. The HE persisted in the area around 42° N, 148° E up to the end of January of the next year. It splitted
 ¹⁰ eventually on January 31, 2012 into two anticyclones.

3.3 The Tsugaru eddy

The anticyclonic TsE was born on February 4, 2012 after splitting decay of the HE (the yellow patch with the elliptic point at its center around , . The core of that

- ¹⁵ eddy 42° N, 145.6° E in Fig. 4a). The elliptic point at the center of the TsE appeared at 41.8° N, 146.9° E. Just after its birth, the HE begun to transport its "yellow" water around the TsE with the core consisted of an Oyashio "bluewater" with a comparatively small amount of the
- ²⁰ Fukushima-derived "yellow water" at its periphery." water (Fig. 4a). Then it gained some modified subtropical "red water" (b). The strong Subarctic Front is visible in Fig. 4 b) and as a contrast boundary between Oyashio "blue" water and Fukushima-derived "yellow" water with the Tsugaru ²⁵ "blackwater" (" water in between.

Seawater samples for radiocesium measurements in the frontal area have been collected during the R/V "*Mirai*" cruise from January 31 to February 5, 2012 along one of observation lines of the World Ocean Circulation

- ³⁰ Experiment (WOCE) in the western Pacific, specifically the WOCE-P10/P10N line (Kumamoto et al., 2014). We impose on the simulated Lagrangian map in Fig. 4c). It was a comparatively large mesoscopic eddy around 150 km in diameter with the elliptic point at a locations of stations
- ³⁵ to the north of the Kuroshio Extension (>36° N) with measured levels of the cesium concentrations. As before, the green diamonds mark locations of those stations, P10-114 (42.17° N, to-143.8° E), P10-112 (41.75° N, 144.13° E), P10-110 (41.25° N, 144.51° E), P10-108 (40.76° N,
- ⁴⁰ 144.88°E), P10-106 (40.08°N, 145.37°E) and P10-104 (39.42°N, 145.85°E), where the cesium concentrations in surface seawater samples have been measured by Kumamoto et al. (2014) to be at the background level.

The stations, P10-102 (38.75°N, 146.32°E), P10-100

- $_{45}$ (38.08° N, 146.77° E), P10-98 (37.42° N, 147.2° E), P10-96 (36.74° N, 147.63° E) and P10-94 (36.08° N, 148.05° E), where the concentrations have been found to be larger (but not exceeding 25.19 ± 1.24 mBq/kg for 137 Cs), are indicated by the magenta diamonds. It's worth to stress a
- ⁵⁰ good qualitative correspondence with our simulation results 10 months after the accident in the sense that stations with measured background level are in the area of Oyashio "blue" waters with low risk to be contaminated, whereas

stations with comparatively high levels of radiocesium concentrations are in the area of the day of sampling on Fukushima-derived "yellow" waters with increased risk of contamination.

As to the TsE, it was sampled later, in July 5, 2012, in the cruise of the R/V "*Professor Gagarinskiy*" (Budyansky et al., 2015) when it was a comparatively large mesoscale eddy around 150 km in diameter with the elliptic point at 41.3° N, 147.3° E consisting of intermittent strips of "blue" and "yellow" waters (Fig. 4d) –

which have been wrapped around during its growth from February to July 2012. Station 84 in that cruise was located near the elliptic point of that eddy (called as G by Budyansky et al. (2015)). The concentration concentrations of ¹³⁷Cs at the surface and at 100 m depth has have been measured to be 11 ± 0.6 Bq m⁻³ mBg/kg and 18 ± 1.3 Bq m⁻³ mBg/kg, respectively, an order of magnitude larger than the background level. As to the ¹³⁴Cs concentration, it was measured to be smaller, 6.1 ± 0.4 Bq m⁻³ mBg/kg and 10.4 ± 0.7 Bq m⁻³ mBg/kg due to a shorter half-lifetime of that isotope. In fact, it was one of the highest cesium concentrations measured inside all the eddy features sampled in the cruise -15 months after the accident.

The maximal concentration of radionuclides 15 months after the accident was observed, as expected, not at the surface but within subsurface and intermediate water layers (100–500 m) in the potential density range of 26.5–26.7 due to a convergence and subduction of surface water inside anticyclonic eddies. The corresponding tracking map in Fig. 10c by Budyansky et al. (2015) confirms its genetical genetic link with the TE, and, therefore, a probability to detect increased cesium concentrations was expected to be comparatively large. We were able to track all the modification of the TSE up and its death on April 16, 2013 in the area around 40° N, 147.5° E.

4 Conclusions

We elaborated a special Lagrangian methodology for simu- 30 lating, tracking and documenting origin and history of water masses in ocean mesoscale features. Integrating advection equations for passive particles in the AVISO velocity field backward in time, we have computed Lagrangian maps demonstrating clearly by which waters the mesoscale eddies 95 in the Kuroshio-Oyashio confluence zone were composed of. It allowed to simulate by which ways they gained and lost water with a risk to be contaminated by Fukushima-derived radionuclides. We have studied three genetically-linked persistent mesoscale anticyclonic eddies in the area, TE, HE and 100 TsE, which have been sampled in the R/V cruises in 2011 and 2012 to contain higher concentrations of radiocesium isotopes. The simulated Lagrangian maps allowed to document and analyze how they interact to pass radioactive water to each other. The simulated results have been shown to be in 105 a good qualitative correspondence as compared with *in situ* measurements.

We hope that the proposed methodology could be applied to simulate propagation of pollutants after future possible ac-

⁵ cidents and identify and track contaminated persistent features in the ocean. The Lagrangian methodology seems to be useful, as well, to plan courses of the R/V cruises. It allows not only to track mesoscale eddies in the studied area but to identify the origin of water masses and to estimate
 ¹⁰ a priori concentrations of radionuclides, pollutants or other

Lagrangian tracers inside the eddies planned to be sampled.

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