- We would like to thank the 2 reviewers for their helpful comments. The manuscript has been revised according to the referees' comments.

Anonymous Referee #1

This manuscript presents a new development of the FLEXPART model to use the mesoscale AROME model. The main interest is the detailed attention paid to turbulent mixing and the development of schemes to correct problems that many users may not even be aware of. The work is thorough and useful, I am happy to recommend publication.

The main comment I would have concerns the treatment of mixing in convective clouds. Most of the paper appears to be written without this in mind, but some sections seem to suggest it is a dominant factor. Maybe this can be clarified in the text with an expanded discussion and some comparisons. Given the importance of clouds, should model comparisons not be shown with and without clouds?

- The FLEXPART model includes a deep convective scheme that redistributes the particles vertically based on the parameterisation by Emanuel and Živković-Rothman (1999). This scheme was not used in our simulations because at a resolution at 2.5x2.5km^2, one can assume that the vertical transport is resolved by AROME. The mixing in convective clouds is treated, to some extent, in FLEXPART-AROME by using the TKE fields from AROME that includes the turbulent mixing from deep and shallow convection. Although mixing in convective clouds is a major advantage of the novel turbulent scheme compared to the old, it was not the main focus of the study. Since AROME is an operational model, it is not possible for us to do an intensive comparison between simulations with and without clouds.

Minor comments:

The captions of Figures 4 and 5 could be expanded. The explanation of the figures in the text could also be expanded for clarity.

- The manuscript has been changed.

I was not sure what the purpose of giving the input codes in Table 1 is. Maybe some useful information can be given to help the reader keep in mind the difference between the schemes?

- The main purpose of Table 1 is to help future users of FLEXPART-AROME to navigate the different turbulent options. The table has been moved to the annexes since it contains no added value to the main text. Thank you for this remark.

Page 7, Line 5: The explanation of bottom-up and top-down could be made clearer. I even wonder about the terminology – maybe a better name could be found for these, and for the turbulence scheme. Section 3.3: As for the comment regarding bottomup/top-down, the turbulence schemes could be better explained.

- We have changed the terminology and altered the discussion to better express the difference between both configurations.

Writing comments:

- Thank you for your thoroughness, the corrections have been taken into account.

Anonymous Referee #2

The manuscript by Verreyken and co-workers presents the development of a novel version of the widely-used Lagrangian particle dispersion model FLEXPART for the use with output from the limited area model AROME. However, the focus of the manuscript is on the development and validation of new turbulence schemes that could be used as an alternative to the default FLEXPART parameterisation that is not building on the turbulence information available from the driving meteorological model. The design and implementation of new turbulence schemes for FLEXPART certainly is an important development and may lead to improved dispersion simulations in many different areas of atmospheric transport. However, I feel that the study was not carried out with the required care and thoroughness and needs major revisions before it can be accepted in GMD. A detailed list of my concerns follows.

Major comments:

Validation:
A number of different new turbulence schemes (or various settings for these) were tested for conservation of well-mixedness and for two different case studies with surface releases. Although, the well-mixedness test is very important, it alone does not seem to be sufficient to judge which setup performance best. The surface release cases remain very qualitative in their evaluation. I had hoped for a more quantitative validation of the turbulence schemes, either by application to existing tracer release experiments or, if this is not easily possible, by an application to real-world observations made at the Maido observatory on La Reunion. The manuscript mentions observations of water vapor isotopes at the observatory, but surely there are also other observed tracers that could be used to identify the PBL influence at the site and could be compared to different transport simulations in a more quantitative way and under different mixing conditions (if possible). Such an analysis is hinted to at the end of the conclusion as part of future work, but I strongly feel that the current manuscript requires this more quantitative validation as well. Without such an analysis I don’t think a clear conclusion can be drawn in terms of which turbulence scheme should be used in future applications and if the new schemes are even performing correctly at all.

- The principle of the turbulent scheme developments for FLEXPART-AROME was to improve consistency between the NWP and the offline LPDM dynamics. The focus of our development efforts was thus numerical consistency when looking at the well-mixedness criterion and the surface release test. We assume that any further testing would not probe the FLEXPART-AROME developments but rather the differences between the online and offline turbulent parametrizations of AROME and FLEXPART respectively.
- Furthermore, existing tracer release experiments were not conducted within any available AROME domains. Characterising the influence of the PBL development at Maïdo observatory on observations is an ongoing research project which will also rely in part on FLEXPART-AROME and the Meso-NH mesoscale simulations during the intensive observation period of the OCTAVE project (March to May 2018). We think that an analysis of the influence of PBL development on Maido observations to quantitatively evaluate the new and old turbulent schemes in FLEXPART will be addressed by the results of the OCTAVE project and will merit a separate publication rather than serve as a validation of the current presented work.
Although it is correct that the well-mixedness criterion on its own does not select the 'best' setup, it does confirm the plausible use of different configurations. It is however clear that the traditional turbulent time step configuration is incompatible with the TKE field ingestion. The choice of L_w parametrisation depends on which one is used in the NWP, the whole point is to get consistent results between the Eulerian and the offline model so it does not make sense to choose one offline, independent of the NWP. Concerning the Step TKE or SDA configurations, users are free to choose which one suits their needs best. One is not 'better' as the other. In general, when users are interested

in a vertical output from FLEXPART-AROME that has a resolution similar or larger than the NWP we suggest using the SDA configuration with elevated values of CTL and IFINE. If the outgrid vertical resolution is lower than that of the NWP we suggest using the Step TKE configuration with slightly lower values of CTL and IFINE to speed up the simulation. The values of these input parameters are essential to the validity of the small discontinuity approximation.

Structure and Presentation:
It is not always easy to follow the flow of the manuscript. It is often not well explained why and how certain things were done (see examples below). In other sections the manuscript is lacking the degree of detail that is important for a model development paper. Also, the current conclusions are lacking a clear recommendation, which of the new turbulence schemes should be used in future studies, and if the developments presented here will make it back into the main and/or WRF FLEXPART versions.

- We have added details and cleared up things as much as possible. There is not a single turbulent setting which can be systematically recommended (a fact that is also true for parameterizations available in mesoscale models) but rather four possible options from which the user should choose depending on the configuration of the NWP, the desired vertical resolution of the output and of the CTL and IFINE parameters as discussed above.

<u>Minor comments:</u>
Abstract, last sentence: This was said before. Maybe reformulate to make it the main conclusion of the study. Also include a statement/recommendation of the default turbulence scheme to be used with FLEXPART-AROME.

- Thank you for the suggestion, we changed the last part of the abstract.

Introduction: The pros and cons of Lagrangian versus Eulerian models are stated. However, this section is lacking good citations and it is also a bit too negative about Eulerian models. For example advection schemes in Eulerian models can be designed in ways that they are conserving mass and are less diffusive. But usually this comes with a prize of higher complexity and larger computational costs. But generally the statement that Eulerian models cannot do this is not valid. Also, there are other uncertainties connected to offline Lagrangian transport models. 1) temporal resolution of input meteorology, when running off-line, 2) less explicit description of turbulence (in comparison to prognostic TKE in most NWPs), exactly what the manuscript highlights later.

- We have reduced the imbalance between Eulerian and Lagrangian models, added a reference and reorganised the manuscript to highlight the difference in turbulent parameterizations in the introduction.

P2,L6ff: Regional inverse modelling studies are also an increasingly important field of application of LPDMs: eg. Stohl et al. (2009), Lin et al. (2003), Manning et al. (2003)

- Thank you for this note, it has been incorporated in the manuscript.

P2,L11f: Does this sentence still refer to different FLEXPART versions? Please mention these again with reference. Next to FLEXPART-WRF, there is also the FLEXPARTCOSMO version mentioned in Pisso et al. (2019) and described in Henne et al. (2016).

- The manuscript has been adjusted.

P2,L15: "French metropolitan area" Does this refer to Paris or the whole mainland France domain?

- The French metropolitan area refers to the whole mainland of France. It is clarified in the new version.

P2,L20: Is AROME-SWIO also an operational model product by MeteoFrance?

- AROME is operationally used over the South-West Indian Ocean by Meteo-France. It is referred to in as AROME-IO or AROME-SWIO. Labelling issues aside, it is an operational model product of Météo France. I've adjusted the manuscript to make clear that it concerns an AROME configuration in the SWIO area to avoid the explicit labelling.

P2,L24: Reference to FLEXPART-WRF publication missing. On which FLEXPARTWRF version is FLEXPART-AROME based?

- FLEXPART-AROME is based on FLEXPART-WRF version 3.1.3.

P3,L9f: The sentence is incomplete. I guess it should continue after (fig 1) without starting a new sentence.

- Indeed, thanks for the remark.

PBL diagnostics: Why is the PBL height diagnostic, which was solely based on theta_v, called robust as compared to the Richardson bulk number approach in FLEXPART? The latter is also using the theta_v profiles from AROME, but in addition it also uses wind shear information (again from AROME). Compare Stohl et al. (2005). However, FLEXPART in its original version uses an "enveloping" PBL height, which is the maximum from the neighboring grid cells and the two model time steps in memory. It also extends the PBL height in areas with large subgrid-scale orographic variability. Both approaches may NOT be justifiable for high resolution simulations and may be the reason for the "overestimation" of PBL heights by FLEXPART in mountainous terrain (P3,L11). Which approach was followed in FLEXPART-AROME? The same as in FLEXPART ECMWF? It is also not clear how PBL heights were estimated solely based on theta_v. By a parcel method? Assuming that the PBL height is the height where theta_v is first larger than theta_v at the surface including a surplus surface temperature? Was an additional interpolation between model levels used (like in FLEXPART)?
- The virtual potential temperature method is called robust since it is a simple and straightforward diagnostic, not to contrast with the parametrisation in FLEXPART. The Richardson bulk number approach in FLEXPART is one of the many possible PBL height parametrisations. We do not want to say that one is better or worse than the other, rather that there is a problem in the number of ways that can be used to determine the PBL height. As an illustration we used a simple, straightforward and robust diagnostic to estimate the PBL height to highlight the fact that PBL tops can differ between parametrisations. If the offline diagnostic is in disagreement with the NWP turbulence it will misrepresent transport of air masses near the top of the boundary layer as is stated in the paper. By using the TKE from the model we no longer depend on offline parametrisations and improve consistency between the models which is the main goal of the development presented here.
The PBL height from FLEXPART as shown in the manuscript does use the 'enveloping' scheme but the subgrid-scale orographic impact is switched off by putting the LSUBGRID input parameter to zero as it indeed is not justified in AROME grid resolution.
- The PBL height based on theta_v is determined at the level where the virtual potential temperature equals the one at the surface with a certain surplus. There is no interpolation between model levels used.
- We have adapted the discussion to a simpler comparison between the FLEXPART boundary layer height ant the TKE fields from AROME for simplicity's sake.

Comparing turbulent layers and PBL heights: The TKE layer diagnosed from AROME is strictly speaking not the same as the classical PBL height. Hence, I suggest to clearly separate the naming from what is otherwise called PBL height. This is implicitly introduced in the description, but it would be better to clearly distinguish between this turbulent layer and the PBL! It would be good to clearly define this layer in section 2 and explain in more detail how it was diagnosed from the model output. Currently this is only done in the caption to Figure 2 although the resulting layer depth is already displayed in Figure 1. From this it is also clear that one cannot conclude from the comparison between turbulent layer and PHL heights that the latter is under- or overestimated (last sentence section 2). One can only say that the former is greater or smaller than the other.

- To avoid confusion, we changed the discussion where we only compare the PBL height from the FLEXPART parametrisation with the TKE fields, as mentioned above.

P3,L15: In FLEXPART it is also possible for particles to cross from the PBL to the FT through the subgrid-scale convection scheme. Was this switched on or off for FLEXPART-AROME. The scale would probably call for switching it off but this was not clearly stated anywhere?

- The FLEXPART subgrid scale convection concerns deep convective motions. This is resolved at the AROME resolutions and is turned off by setting the LCONVECTION input parameter to zero.

P3,L21: This "erratic behavior" could be avoided by detecting the layer top where at least two neighboring levels show TKE below the threshold value. From the examples given in Fig 2. It seems it is always a single level with low TKE between PBL and shallow convection zone. Does the erratic behavior actually matter for the TKE-based turbulence scheme in FLEXPART-AROME? Or is it only a diagnostic for the comparison with FLEXPART's PBL height estimation? This should be clarified in the text as well.

- The height of the turbulent layer in AROME does not influence the transport in FLEXPART-AROME but is rather a diagnostic from AROME output most comparable to the PBL height, which is why it was shown here. In FLEXPART-AROME the turbulent layer height is not used in any way.

One last question concerning the TKE layer height: From the layer heights displayed in Fig. 1, my conclusion would be that mixing over the sea is more intensive or at least reaches higher than over the mountains. This is counter-intuitive, but possibly related to the fact that heights above ground are shown. What are the model orography heights for the 4 points for which the layer heights were evaluated in Fig 1?

- The surface level in the mountains is 1222 m above sea level. The counter-intuitive conclusion that mixing above sea reaches higher altitudes is indeed due to the fact that heights are shown in meter above ground level.

P5,L15: At this point not clear what a Thomson interface is and there is no reference given either.

- This is indeed a specific term that is not referenced anywhere else in the paper. It is a name we use for the interfaces at which the particles can reflect and is described elsewhere in the manuscript as 'TKE interfaces'. We have changed this here.

P5,L15 and section 3.1: How does the Thomson approach actually justify setting the density correction to zero? The density is not affected by turbulence intensity and as such density gradients are not explicitly treated by the Thomson approach.

- The density correction is not set to zero, only the drift correction. Thank you for the remark. As said in section 3.1, Lin et al. (2003) did include a density correction in their implementation of the method proposed by Thomson et al. (1997) but we opted to keep the FLEXPART density correction. Both possibilities were tested and had identical results. We decided to keep the FLEXPART density correction implementation.

P5,L18ff: Point out that this choice refers to the 1D, 3D options in Table 1. Please give a reference to the "diagnostic equations from Meso-NH" so that these can be found by the interested reader or even repeat them here if they are central.

- Turbulent motions implemented in Meso-NH can be found in Cuxart et al. (2000). They are not central since they are not specifically tested in this work. We have added the reference. Thank you for the remark.

P7,L3ff: The two different ways how to calculate the time step should be introduced much clearer and the terms 'bottom-up' and 'top-down' properly introduced as two ways how to calculate the turbulent time step. These terms have different meanings in different fields and in the context of the time step it remains a bit unclear why they were chosen.

- We have rewritten this part and changed the terminology to adaptive vertical turbulence time step (FVTTS) versus a fixed vertical turbulence time step (AVTTS).

Turbulent mixing length: Would it be possible to give the equations for the three different ways how L_w was calculated?

- We have added the implementation of these parametrisations in the annexes of the manuscript.

Section 4, Validation: The setup should be described with more care and detail. For example: Were mean wind fields set to zero for this case? Were actual fields from AROME used for this exercise or some standard fields? Does it matter which exact locations were chosen? The location should not matter, only the surface properties, if winds were set to zero. How many particles were used in these exercises? How many per grid column? What was the FLEXPART PBL height in these grid cells and how did the TKE profile look like. Maybe both could be added to Fig 4.

- Vertical turbulent motions were isolated by taking out displacements along the resolved winds. The TKE fields used for the tests are 3D fields obtained from AROME. The tests were run in a single column with 250000 particles. Different locations give similar results but since the TKE profiles differ between grid cells we specified which grid cell was chosen for the tests. A figure showing the TKE profiles and the PBL top evolution was added to the manuscript.

Section 4: The 3D configurations of the turbulence scheme are never discussed only introduced in section 3. Were these schemes not tested after all? If not, why introduce them in the methods sections? If they were tested, how did the results compare to the 1D cases?

- The 3D turbulent modes were not validated as the 1D modes as we have no AROME runs with 3D turbulence. These modes were implemented to anticipate future developments and were only checked to not provide problems when running the software.

P8,L26: 'TURB_OPTION=11 and 111'. In the previous sentence the settings were spelled out not just the option index given. To keep the text flowing the same should be done here. The option index could still be given in braces (wherever this helps to clarify things). The sentence is also a bit odd, because already the previous sentence said 'DEARDORFF [...] has the least accumulation', which is the same as ’best conserve well-mixed state’. Better start with ’Besides the DEARDORFF modes, modes xxxx best coserve ...’.

- Thank you for the remark, we have adjusted the manuscript.

P8,L29ff: The discussion on L_w could be more illustrative if profiles of L_w could also be included in Fig 4 or together with PBL heights and TKE profiles in a supplement.

- We agree that showing values of L_w would be more illustrative. Unfortunately, since this is a local value that is recalculated at every step for each particle we cannot retrieve this kind of profile.

Figure 4/5: Why is the second WRF TKE mode called ’stable repartitioning’ here? It was introduced as a ’TKE only’ method above. No mention of it being stable. In the caption it should be repeated that these are results for a grid cell over the ocean.

- The second WRF TKE method is indeed based only on the TKE value without the PBL parameterisation used in FLEXPART. The stable repartitioning refers to the parametrisation used by FLEXPART-WRF to distribute the TKE over 3 dimensions.

P9,L4: I think this should be ’Near-surface concentrations".

- Indeed, thank you.

Section 4, Fig 5: How is it possible that the vertical gradients in the simulations with new turbulence scheme are maintained over time. Even if the mixing in the shallow convection zone is smaller than in the PBL, one would expect that the vertical gradient eventually vanishes in the 24 hours of simulation, as it quickly does in the Hanna case within the PBL. In order to illustrate this more clearly, it would also be interesting to see the final mixing ratio profiles of all configurations in a comparison.

- The gradients are maintained over time due to the use of dynamic TKE profiles obtained from AROME. At certain times mixing reaches higher altitudes after which these particles are not mixed further. We have added final mixing ratio profiles in an annex as it is indeed interesting to see but does not contain new information compared to the plots shown in Figure 5.

Section 5: It would be valuable if this section would show some kind of comparison with observations at Maido. Such a comparison could help supporting the authors suggestion that the new turbulence modes are superior to previous schemes. Without such a comparison there is little evidence that the performance of the new schemes is more realistic.

- As stated before, the idea of these new developments were to get consistency between the offline transport model and the NWP. We don’t claim that one is superior to the other but rather that we should not have a difference between the online and offline turbulent parametrisations for the reasons mentioned above.

P10,L3: It is always confusing with LPDMs to write about ’particle transport’ which can easily be confused with aerosol particle transport. Here, it would probably work best to simply replace ’particle transport’ by ’atmospheric transport’.

- Thank you for the suggestion, we have changed the wording.

Figures 6/7: Most of the text of the figure captions is also stated in the main text. Remove from caption. Rather repeat what is seen in each sub-panels. Sub-panels should also be labeled by letters.

- The manuscript has been adapted.

Section 5.2: This section needs some introduction on how computation times were estimated. Were repeated runs carried out for each configuration? This is important since run-times may differ due to other processes running on the same machine and/or I/O may be influenced by other processes. It would also be helpful to mention on what architecture and with which compiler (options) and with which parallelisation approach these results were obtained. Do these timings reflect run times for the complete model runs or just for the transport part of the model? Please speculate why the increase in computation time was so much larger for the well-mixed test compared to the surface release? Why were computation times larger for the no-turbulence cases? Shouldn't these perform much faster, since only the mean motion needs to be solved for (which was zero in the well-mixed and point release tests)? I see it is explained later on. So only a quick-and-dirty implementation of no turbulence was used. But then one should not compare these run-times. There is little value in it since they present some kind of artificial, never-used option.

- No repeated runs were carried out for each simulation as the simulations were run on a table-top machine which was dedicated to the FLEXPART-AROME simulations during the test phase. Simulations were run on a workstation with a single CPU INTEL CORE I7-7700, 32 Gb of DDR4 SDRAM with a GNU compiler. The machine was dedicated to the FLEXPART-AROME simulations to minimise the impact of parallel processes on the computation times. Currently the code is not parallellised but it is foreseen to update this and to use the openMP approach similar to the FLEXPART-WRF code. Run times reflect the complete model runs. We agree that the implementation of no-turbulence is quick-and-dirty. However, since it is the way it is implemented in the FLEXPART-WRF code and it is a possible choice of input by the user, we decided to include it in the comparison.

Conclusions: These are a bit non-conclusive. So what is the recommendation for future use of the model? Which turbulence mode should be used and why?

- We have adjusted the manuscript to reflect the considerations that were addressed in previous answers.

P13,L6: Shouldn't this be 3D TKE fields? Which dimension would be dropped out for them to be 2D?

- Indeed, this is a typo. Thank you.

<u>Technical comments:</u>

- Thank you for the comments, it has been fixed.

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Development of turbulent scheme in the FLEXPART-AROME v1.2.1 Lagrangian particle dispersion model

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Abstract. The FLEXible PARTicle dispersion model FLEXPART, first released in 1998, is a Lagrangian particle dispersion model developed to simulate atmospheric transport over large and meso-scale mesoscale distances. Due to FLEXPART's success and its open source nature, different limited area model versions of FLEXPART were released making it possible to run FLEXPART simulations by ingesting WRF (Weather Research Forecasting model), COSMO (Consortium for Small-scale Modeling) or MM5 (meso-scale mesoscale community model maintained by Penn State university) meteorological fields on top of the ECMWF (European Centre for Medium-Range Weather Forecasts) and GFS (Global Forecast System) meteorological fields. Here, we present a new FLEXPART limited area model that is compatible with the AROME mesoscale meteorological forecast model (the Applications of Research to Operations at Meso-scale Mesoscale model)[1]. FLEXPART-AROME was originally developed to study meso-scale mesoscale transport around La Réunion, a small volcanic island in the South West Indian Ocean with a complex orographic structure which is not well represented in current global operational models. The AROME vertical hybrid sigma grid is projected on the Cartesian terrain following FLEXPART grid. We present new turbulent modes in FLEXPART-AROME. They differ from each other by: dimensionality, mixing length parameterisationparameterization, turbulent transport constraint interpretation and a novel time-step configuration. Performances of new turbulent modes are compared to the ones in FLEXPART-WRF by testing the conservation of well-mixedness by turbulence, the dispersion of a point release at the surface and the marine boundary layer evolution around Reunion island. An adaptive time step for the vertical turbulent motions has been implementedto improve conservation of well-mixedness in the modelThe novel time-step configuration proved necessary to conserve the well-mixedness in the new turbulent modes. An adaptive vertical turbulence time step was implemented, allowing the model to adapt on a finer time scale when significant changes in the local turbulent state of the atmosphere occur..

[1]Applications de la Recherche à l'Opérationnel à Méso-Echelle

1 Introduction

Atmospheric transport models are divided into Eulerian and Lagrangian transport models. Eulerian models represent the atmosphere in a grid with mass being exchanged between grid cells. They are especially useful to model chemical interactions in the atmosphere. However, Eulerian models are unable to maintain have difficulties maintaining the shape of narrow plumes due to numerical diffusion in their advection scheme. A number of techniques can be applied to dampen these diffusions but they generally come with great computational costs (Alam and Lin, 2008). The Lagrangian models on the other hand describe the evolution of air masses in pregenerated 3D wind meteorological fields obtained from a numerical weather prediction (NWP) modelallowing precise , allowing precise and fast modelling of atmospheric tracers released from point-sources. Uncertainties in Lagrangian models are limited and originate from naive originate from linear temporal and spatial interpolation from the 3D meteorological fields of the NWP model (Stohl et al., 1995). Lagrangian particle diffusion dispersion models (LPDM) such as FLEXPART represent an air mass by a large amount of infinitesimally small air parcels, also called particles, into the atmosphere. Each individual particle is advected along the resolved wind fields with a turbulent diffusion superimposed. (Zannetti, 1990)

LPDMs are used in a variety of atmospheric studies such as source apportionment of chemical compounds (Gentner et al., 2014; Warneke et al.), studying atmospheric water vapor transport (Bertò et al., 2004; D'Aulerio et al., 2005; James et al., 2008), characterising deep stratospheric intrusions (Brioude et al., 2007; Akritidis et al., 2012), as well as hazard preparedness exercises (Stohl, 2013). Regional inverse modelling studies are also an increasingly important field of applications of LPDMs (Lin et al., 2003; Manning et al., 2003; Stohl et al., 2009; Brioude et al., 2011).

Pisso et al. (2019) describe the FLEXPART offline transport model . The latest release ingests meteorological data from the ECMWF and GFS global model . Several limited area models have already been developed allowing , including the available limited area model versions. The limited area versions of FLEXPART (FLEXPART-WRF (Brioude et al., 2013), FLEXPART-COSMO (Henne et al., 2016), FLEXPART-MM5) allow particle transport in higher resolved grids with the possibility to better represent the mesoscale phenomenain the atmospheremesoscale phenomena.

The AROME mesoscale forecast model has been the operation weather forecasting model at Météo France since 2008. It is designed for fine-scale modelling with grid sizes ranging from 0.5 to 2.5 km. AROME is developed by combining efforts of the French Meso-NH research model community and the ALADIN consortium[2]. Since 2015, the French metropolitan area mainland France is covered by a 1.3 km horizontally resolved grid in a Lambert conformal projection which results not only in a more realistic representation of topologically induced physical phenomena but also allows for a fine scale variation in surface types impacting for instance the sensible heat flux at the surface (MétéoFrance). FLEXPART-AROME was developed by the LACy laboratory to model particle transport around La Réunion, a french overseas territory which is covered by an AROME grid in the South-West Indian Ocean (AROME-SWIOSWIO) with 2.5x2.5 km^2 horizontal resolution in a Lambert Conformal projection. With its 90 vertical hybrid sigma levels it reaches an atmospheric altitude of about 24 km above sea

[2]The ALADIN consortium contains the Algerian, Austrian, Belgian, Bulgarian, Croatian, Czech Republic, French, Hungarian, Moroccan, Polish, Portuguese, Romanian, Slovakian, Slovenian, Tunisian and Turkish weather services.

level. A ~~provisionary~~ provisional version of FLEXPART-AROME was successfully used in the 2015 STRAP campaign to forecast transport of a volcanic plume on the Island (Tulet et al., 2017).

FLEXPART-AROME is based on the FLEXPART-WRF v3.1.3 code which is able to use the Lambert Conformal projections in the horizontal coordinate. The hybrid sigma levels are projected on Cartesian terrain-following vertical levels used by FLEXPART. To simulate turbulence induced by the complex orographic structure of the volcanic island of La Réunion and by shallow convection, we built on the turbulent modes implemented in FLEXPART-WRF by ingesting the 3D turbulent kinetic energy (TKE) field from the NWP in FLEXPART in order to harmonise turbulent motions between both.

2 ~~Turbulent inconsistency between NWP and LDPM~~

Incoherent turbulent representations may introduce unrealistic tracer transport features. For instance, if the planetary boundary layer (PBL) height ~~(PBL)~~ is overestimated in the transport model, tracers will be advected along stronger free tropospheric (FT) winds with a different direction. If the reverse is true and the PBL height is underestimated a passive tracer released at the surface will be well-mixed over a smaller vertical range, overestimating tracer concentrations in the boundary layer.

The FLEXPART Lagrangian particle dispersion model uses ~~a turbulent parameterisation independent of the NWP model. It was~~ the turbulent parameterization proposed by Hanna (1982) ~~, developed and validated for meso-scale models. The PBL height is calculated by the method proposed by Vogelezang and Holtslag (1996)~~and computes the PBL top along the method of Vogelezang and Holtslag (1996). In the large-scale global grids, deep convection is a relevant sub-grid scale process. To describe this, Forster et al. (2007) adapted the convective parameterization by Emanuel and Živković Rothman (1999) in FLEXPART. Deep convection is assumed to be resolved in the mesoscale grids from AROME. The scheme was switched off by setting the LCONVECTION input parameter, introduced in FLEXPART-WRF, to zero. FLEXPART-WRF ~~implemented~~ introduced two new turbulent modes using the 3D TKE fields ~~. However, they were~~from the NWP model. They were, however, reported to violate the well-mixedness condition, described by Thomson (1987), which states that ~~turbulent behaviour~~turbulence cannot change an initially well-mixed ~~atmosphere~~atmospheric tracer. To resolve this in the newly implemented turbulent modes in FLEXPART-AROME, we applied the method proposed by Thomson et al. (1997), successfully used in the Stochastic Time-Inverted Lagrangian Transport (STILT) model (Lin et al., 2003), to constrain particle transport ~~keeping with the well-mixed criterion. When comparing the PBL localisation in FLEXPARTwith robust estimates based on the temperature (θ_v) profile in AROME (fig 1). Above sea FLEXPART seems to systematically underestimate the PBL top location while in a mountainous region the reverse is true. When using TKE fields~~ at discrete interfaces in the model.

In contrast tot he Hanna turbulence in FLEXPART, AROME TKE fields include shallow convective transport, allowing novel turbulent modes in FLEXPART-AROME to mix boundary layer air with free tropospheric air masses.

Figure 1 illustrates the difference between the TKE fields from AROME and the calculated boundary layer top[3] from FLEXPART. We note that there is a large difference in ~~AROME to check the depth of the turbulent layer starting from the~~

[3] Subgrid-scale orography variations and enveloping PBL height considerations, that can be taken into account in FLEXPART, are not taken into account since they don't make sense at the current mesoscale resolutions.

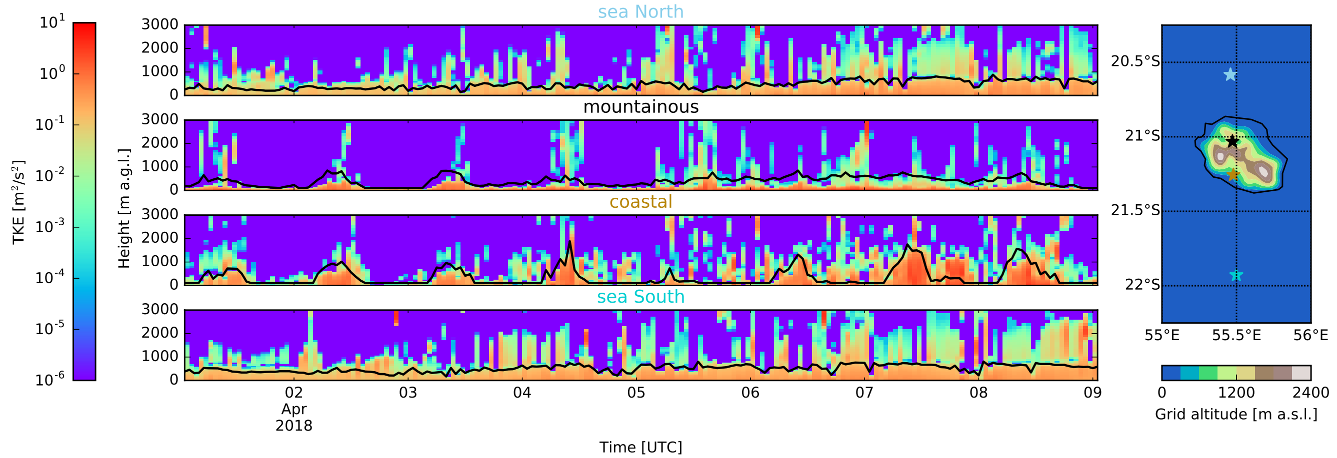


Figure 1. PBL top time Temporal evolution according to FLEXPART (green) and inferred of TKE fields retrieved from AROME in four different types of area around Réunion Island overlayed with a black curve showing the θ_v profiles PBL top as calculated in AROME (red)FLEXPART. Blue represent The vertical evolution plots on the turbulent layer left correspond from top to bottom with the surface up including turbulent clouds resolved in locations indicated on the tke profile map from AROME. Shaded background indicate local night timeNorth to South respectively. Based on TKE profiles, the turbulent region at Height of the surface vertical profiles is deeper than expressed in meter above ground level, over the PBL top suggests. Differences between parameterisations cause over- or under mixing of surface tracers inducing density variations between independent modelsmountainous and coastal areas this corresponds with and added 1.2 and 0.4 km above sea level respectively.

surface, the comparison is less straightforward. Since turbulent kinetic energy in AROME includes energy from turbulent motions in FLEXPART-WRF modes, where turbulence is only treated within the PBL, and the turbulent kinetic energy fields retrieved from AROME. The inclusion of shallow convection and convective clouds, clouds situated at the PBL top allow surface tracers to cross the PBL topinto the free troposphere. This last is a major difference between the novel TKE formalism in FLEXPART-AROME and FLEXPART turbulence. In FLEXPART, mass transport between PBL and the FT is only possible by resolved winds and particles reaching the PBL top by turbulent motions are reflected. In FLEXPART-AROME we do not define the PBLregion and only look at turbulent versus non-turbulent regions. Convective clouds reaching and crossing the PBL top are simply treated as turbulent regions promoting mixing not explicitly represented in FLEXPART. This is clearly illustrated in the vertical profiles of θ_v and TKE, shown in figure ??. It is also clear in this figure that due to the arbitrary nature of the tke limit used in diagnosing turbulent layer depth in AROME, small variations in bridging the turbulence between PBL and clouds can cause what seems erratic behaviour in the diagnosed turbulent layer depth. On average, FLEXPART overestimates the PBL top over land by 133 m while over sea it is underestimated by 158 m (a difference in PBL depth of +25% and -61% respectively) over the 9 day period we randomly selectedconvective clouds in the TKE fields will allow particles at the surface to mix to higher altitudes in the atmosphere.

~~Vertical cross sections for three subsequent hours above the sea South of Reunion Island. Red shows the θ_v profile, blue shows the TKE profile. Horizontal lines correspond with PBL heights from FLEXPART and θ_v, shown in green and red respectively. The blue horizontal line characterises the turbulent layer from the surface including convective clouds based on the TKE profile. The turbulent layer top is defined as the lower bound of the model layer where the TKE drops below 1.e-4 m^2/s^2. A fast inclination of θ_v indicates the beginning of the FT, we found that the altitude where θ_v is 0.5K above its surface value robustly corresponds with the PBL top.~~

2 Turbulent scheme development

Table 1. ~~Different turbulent options introduced in FLEXPART-AROME and their configuration.~~

~~TURB_OPTION~~		~~AVTTS~~		~~FVTTS~~	
		~~1D~~	~~3D~~	~~1D~~	~~3D~~
~~Step TKE~~	~~DELTA~~	~~10~~	~~15~~	~~20~~	~~25~~
	~~BL89~~	~~11~~	~~16~~	~~21~~	~~26~~
	~~DEARDORFF~~	~~12~~	~~17~~	~~22~~	~~27~~
~~SDA~~	~~DELTA~~	~~110~~	~~115~~	~~120~~	~~125~~
	~~BL89~~	~~111~~	~~116~~	~~121~~	~~126~~
	~~DEARDORFF~~	~~112~~	~~117~~	~~122~~	~~127~~

Turbulence in FLEXPART and FLEXPART-AROME is assumed Gaussian and parametrised using a Markov process to solve the Langevin eqation. For a discrete time step implementation this results in:

$$\left(\frac{w}{\sigma_w}\right)_{k+1} = r_w \left(\frac{w}{\sigma_w}\right)_k + \sqrt{1+r_w^2}\zeta + \frac{\partial \sigma_w}{\partial z}\tau_{L_w}(1-r_w) + \frac{\sigma_w}{\rho}\frac{\partial \rho}{\partial z}\tau_{L_w}(1-r_w), \tag{1}$$

where ~~w~~ w is the vertical wind component of the turbulent motion, L_w the turbulent mixing length, τ_{L_w} the Lagrangian time scale for the vertical autocorrelation, σ_w the vertical turbulent velocity distribution width, ρ the air density, ~~z~~ z the altitude, ~~$r_w = \exp{-dt/\tau_{L_w}}$~~ $r_w = \exp\left(-dt/\tau_{L_w}\right)$ the autocorrelation of the vertical wind and ζ a normally distributed random number with mean zero and unit standard deviation. The subscript k and $k+1$ refer to subsequent times separated by dt. The first two terms on the right hand side represent the native autocorrelated turbulent velocity behaviour. The third and fourth terms represent drift and density corrections respectively.(Stohl et al., 2005)

To determine τ_{L_w} and σ_w, FLEXPART-WRF has four modes defined by the TURB_OPTION input parameter introduced by Brioude et al. (2013):

- TURB_OPTION = 0: Turbulent velocities are set to zero.
- TURB_OPTION = 1: Turbulence is computed using the standard FLEXPART configuration using the ~~parametrisation~~ parameterization proposed by Hanna (1982).

– TURB_OPTION = 2: A hybrid configuration combining TKE fields from WRF and FLEXPART parametrisationthe FLEXPART parameterization. Surface-layer scaling and local stability with the Hanna scheme determine the 3D partitioning of the turbulent kinetic energy.

– TURB_OPTION = 3: Turbulent motions are characterised directly by the TKE field from WRF and 3D partitioning is based on balancing production and dissipation of turbulent energy.

Brioude et al. (2013) reported spurious accumulation when using modes where TKE fields from the WRF are taken to characterise the turbulence.

In the FLEXPART-AROME code, the density and drift correction is set to zero and replaced by using Thomson interfaces. The turbulent configurations are also the numerical method discussed in section 2.2. Turbulent modes are extended by 24 modes summarised in table A1configurations. We separated the new options according to the characteristics of each mode, these characteristics will be discussed in greater detail below. The user has a choice in parametrisations for mixing length, the time-loop configurationand the partitioning of TKE, the computation of local TKE and parameterizations for mixing length. Turbulent motions can be restricted to the vertical axis (1D), as it is in AROME-SWIOthe AROME configuration over the SWIO, or partitioned in 3D using the diagnostic equations from Cuxart et al. (2000), implemented in the Meso-NH (Lac et al., 2018) mesoscale model. The 3D modes are not explicitly evaluated here but are rather implemented to anticipate future AROME developments and use of the model in combination with Meso-NH simulations resolved on the fine-scale.

The different novel turbulent modes together with their input parameters are summarised in table A1 (Appendix A).

2.1 Particle time loop

FLEXPART discriminates between the particles below, and those above the PBL top. Above the PBL, particles are advanced in one user defined model synchronisation (LSYNC) time step. In the PBL, particle positions are updated along a leap-frog between turbulent transport and resolved wind fields. The Δt timestep, used by the leap-frog, is determined by the atmospheric stability and the user defined input parameter CTL. Vertical turbulent transport is handled in a second IFINE time loop with a time step $dt = \frac{\Delta t}{\text{IFINE}}$, where IFINE is a third user defined input parameter.

A major difference between the FLEXPART-AROME model and other FLEXPART versions is the treatment of turbulence at the PBL top. By direct use of TKE field from the NWP model, we don't characterise the PBL height explicitly. All particles are put through the time loops. In low turbulent regions, σ_w is small which naturally results in longer time steps:

$$\tau_w = \frac{L_w}{\sigma_w}, \qquad \Delta t = \frac{\tau_w}{\text{CTL}}, \tag{2}$$

where L_w is the turbulent mixing length.

Traditionally, dt is fixed over a Δt period. However, in the new turbulent modes from FLEXPART-AROME, TKE can change abruptly, resulting in significant differences between adjacent dt time steps that are not represented. To resolve this, an adaptive vertical turbulence time step (AVTTS) was implemented. The local time step is computed as:

$$dt' = \frac{\tau_w}{\text{CTL} \times \text{IFINE}}. \tag{3}$$

After IFINE displacements, the local dt' steps are accumulated in $\Delta t = \sum_{i=1}^{IFINE} dt'_i$, which is then used as the time step to displace the particle along the resolved winds.

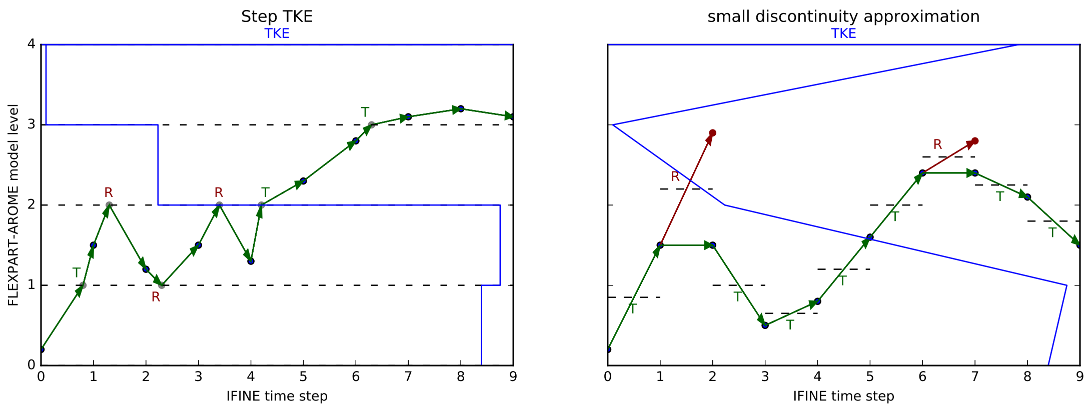
This new time loop configuration is significantly different to the traditional fixed vertical turbulence time step (FVTTS) configuration. As will be shown in section 3.1, the FVTTS is not compatible with new turbulent modes and users of FLEXPART-AROME should always use the AVTTS configuration.

2.2 Thomson's approach

Thomson et al. (1997) discussed the transport of particles through discrete interfaces in a random walk dispersion model. To conserve a well-mixed profile in a turbulent system with discrete TKE steps, particle transport is constrained between different TKE regions. By imposing a net zero mass-flux at TKE interfaces in a well-mixed system and assuming maximal mixing, particles attempting to cross an interface have a probability α of reflection. This probability is proportional to the ratio of Gaussian turbulent velocity distribution widths. Lin et al. (2003) introduced a correction to this probability due to density variations. In FLEXPART-AROME, this correction was not implemented as it is taken into account when solving the Langevin equation (Stohl and Thomson, 1999).

In FLEXPART-AROME, two possible interpretations of ~~this principle~~ Thomson's approach have been implemented. The first considers each displacement a small discontinuity while the second arises from the grid definition of the FLEXPART-AROME model. In the small discontinuity approximation (SDA), turbulent kinetic energy is interpolated in time and space for both the initial, and the final position of a time step dt. The particle is supposed to cross an imaginary interface located at the middle of its trajectory. The probability of crossing is given by $\alpha = \frac{\sigma_f}{\sigma_i}$, where σ_i and σ_f represent the widths of the turbulent velocity distributions at the initial and final position respectively. ~~The difference between both iterpretations is visualised in figure 2.~~ Alternatively, one can consider the FLEXPART grid as a stack of homogeneously turbulent cells. The ~~vertical~~ cell-boundaries are discrete TKE interfaces and particles attempting to cross into an neighbouring cell are reflected with a probability α. In this mode (Step TKE), particles moving a distance dz are checked to see if they cross the cell boundary. If so, the time step is split up in the time it takes for the particle to get to the boundary (dt_1), and the remaining time ($dt_2 = dt - dt_1$). When a particle crosses the boundary, the turbulent velocity is recalculated at the boundary to be consistent with the new local turbulence. The difference between both interpretations is visualised in figure 2.

Both options have their merit. The SDA is recommended when users are interested in a more detailed vertical profile for the FLEXPART-AROME output. Once the SDA mode is selected, users should pay attention to the IFINE and CTL parameters. If



Illustrative difference between Step TKE configuration and SDA. Dashed lines represent TKE interfaces, in the Step TKE configuration they are fixed with homogeneous TKE regions inbetween, the SDA interpolates TKE to the particle position and initialises an imaginary temporary TKE interface halfway the particles trajectory each step. Every time the particle tries to cross an interface we evaluate the probability of crossing and the particle will be either transmitted through (T), or reflected (R) at the interface. The Step TKE configuration updates particle positions to the boundary before computing the probability of crossing (grey points), when particles are transmitted, their turbulent velocity is adapted to the new model layer. The SDA configuration uses a virtual position which becomes reality upon transmission or which is never realised upon reflection (red points).

Figure 2. Illustrative difference between Step TKE and SDA configurations. Dashed lines represent TKE interfaces, in the Step TKE configuration they are fixed with homogeneous TKE regions inbetween, SDA interpolates TKE to the particle position and initialises an imaginary temporary TKE interface halfway the particles trajectory each step. Every time the particle tries to cross an interface we evaluate the probability of crossing and the particle will be either transmitted (T)through, or reflected (R) at the interface. The Step TKE configuration updates particle positions to the boundary before computing the probability of crossing (grey points), when particles are transmitted, their turbulent velocity is adapted to the new model layer. The SDA configuration uses a virtual position which becomes the new position upon transmission or which is never realized upon reflection (red points).

their values are low[4], the small discontinuity hypothesis no longer stands. When users want to speed up their model run and are not interested in detailed vertical distributions near the surface we suggest the use of the Step TKE option.

2.3 Particle time loop

FLEXPART particles are categorised in below PBL and above PBL. Above the PBL, particles are advanced in one user defined model synchronisation (LSYNC) time step. Below PBL the positions are updated along a leap-frog between turbulent transport and resolved wind fields. The Δt timestep is determined by the atmospheric stability and the user defined input parameter CTL.

[4] In our experience, we found that values of IFINE and CTL of 5 were advisable from the different tests. Simulations with CTL values of 2 showed accumulation in all modes, even when combined with IFINE values of up to 10. When using the Step TKE mode modes, we suggest not going to values of IFINE and CTL below 5 and 3 respectively. Our recommendations for the SDA mode are to keep to a minimum of 5 for both parameters.

~~Vertical turbulent transport is handled in a second IFINE time loop with a time step $dt = \frac{\Delta t}{IFINE}$, where IFINE is a user defined input parameter.~~

~~A major difference between the FLEXPART-AROME and other FLEXPART versions is this discrimination at the PBL top. By direct use of TKE field from the NWP model we don't characterise the PBL height explicitly and all particles are put through the time loops. In very low turbulent regions, σ_w is low which naturally results in longer time steps:~~

$$\Delta t = \frac{\tau_w}{\mathrm{CTL}}, \qquad \tau_w = \frac{L_w}{\sigma_w}.$$

~~When large steps in TKE are made by a particle, Δt can change significantly. In the traditional FLEXPART code, this mismatch in time step carries through the remaining IFINE loops resulting in incorrect representation of the turbulent state in the new turbulent modes. In FLEXPART-AROME bottom-up time loop is implemented where Δt is accumulated during IFINE dt time steps where:~~

$$dt = \frac{\tau_w}{\mathrm{CTL} \times \mathrm{IFINE}}.$$

~~In each IFINE loop, this dt is recalculated resulting in an adaptive time step in the bottom-up time loop configuration. Individual particles evaluate their local time step after each displacement and tell the algorithm how long it took them to finish IFINE steps. This in contrast to the top-down FLEXPART implementation where dt is constant throughout a precomputed Δt time step.~~

2.3 Turbulent mixing length

There are currently three ~~parametrisations~~ parameterizations for the turbulent mixing length available in FLEXPART-AROME. The first is based on the grid size (DELTA). It is commonly used as the characteristic length scale of sub-grid eddies and is justified when the grid size falls into the inertial subrange of the turbulent flow and is recommended when the NWP model has high resolution and a nearly isotropic grid (Cuxart et al., 2000). The second ~~parametrisation~~ parameterization is the Bougeaul-Lacarrère mixing length (BL89), a non-local turbulent mixing length ~~parametrisation~~ parameterization proposed by Bougeault and Lacarrere (1989) that balances the TKE with buoyancy effects to determine the mixing length. This ~~parametrisation~~ parameterization is the default mixing length used in the ~~AROME-SWIO model~~ AROME model over the SWIO domain. The last ~~parametrisation~~ parameterization (DEARDORFF) is the analytical limit of BL89 in a stably stratified atmospheric limit which corresponds with the results of Deardorff (1980). It was implemented to study the model behaviour in numerical tests. The use of this last ~~parametrisation~~ parameterization is discouraged for realistic atmospheric transport. The implementation of these parameterizations is discussed in appendix B. Users of FLEXPART-AROME are encouraged to use the same mixing length parameterization as their AROME domain to get consistent results between the NWP and the LPDM.

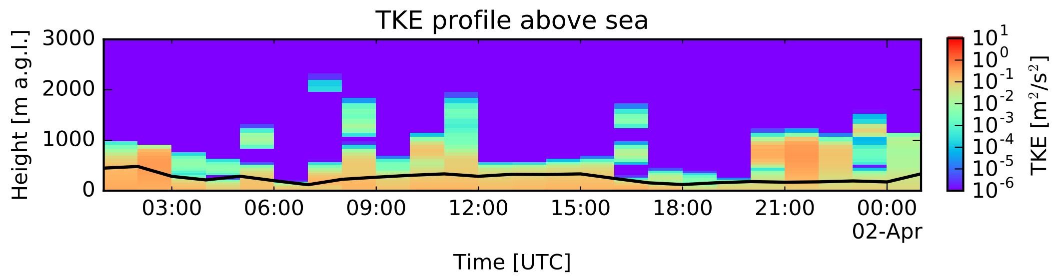


Figure 3. The real TKE pofile used in the validation tests above sea. The black curve corresponds to the PBL height computed by FLEXPART.

3 Validation

Validation tests were run using ~~CTL~~LSYNC=~~5, IFINE~~300, CTL=5 and ~~LSYNC~~IFINE=~~300~~ 5 with output each 30 minutes during a period of 24 hours. For each test 250000 particles are initialised. The particles are not advected along resolved winds to isolate vertical turbulent motions. The horizontal domain is constrained to one ~~AROME-SWIO~~ AROME gridcell area over land or over sea. The output kernel of FLEXPART, spreading a fraction of particle mass over adjacent horizontal cells, was compensated by adding the output between adjacent cells of FLEXPART-AROME output. The grid cells over land and sea were randomly selected to perform our tests. The cell over land has coordinates ~~21.1241S 55.3791~~21.124S 55.379E, corresponding to a forest area on Reunion island. The cell over sea ~~located at 22.4098~~is located at 22.409S 53.939E, a cell 200 km South-West of the island. The vertical output grid goes up to 5km and is resolved by 100 m thick layers. Real TKE fields were used for the test which is why two types of area were explicitly tested. Simulations above sea are shown here, results over land were similar unless explicitly stated otherwise. The TKE profile and the diagnosed PBL height from FLEXPART in the cell above sea are shown in Figure 3

3.1 Turbulent conservation of a well-mixed passive tracer

Initially well-mixed passive tracers in position and velocity space should remain unchanged in a turbulent flow. Isolating the vertical turbulence ~~, by setting 3D resolved winds to zero,~~ and using the MDOMAINFILL option to initialise a well-mixed passive tracer, all turbulent modes in FLEXPART-AROME were tested. Accumulation is normalised to the initial mean mixing ratio. By using the MDOMAINFILL option, numerical fluctuations lead to background accumulations and ~~dilution~~ dilutions of 3.5% and 4.0% respectively. Results above the sea are shown in ~~fig~~ Figure 4.

The Hanna ~~parametrisation~~ parameterization shows systematic accumulation at the surface(11.0%). ~~Modes implemented~~ Turbulent modes introduced in FLEXPART-WRF based on TKE violate consistently the well-mixed criterion~~with turbulent options based on the TKE fields performing worse~~. Dilution at the surface in ~~TURB_OPTION=2 mode being~~ the hybrid FLEXPART-WRF mode is 46.4%~~and~~ , accumulation at the PBL top 42.3%. The results in ~~TURB_OPTION=3~~ the second

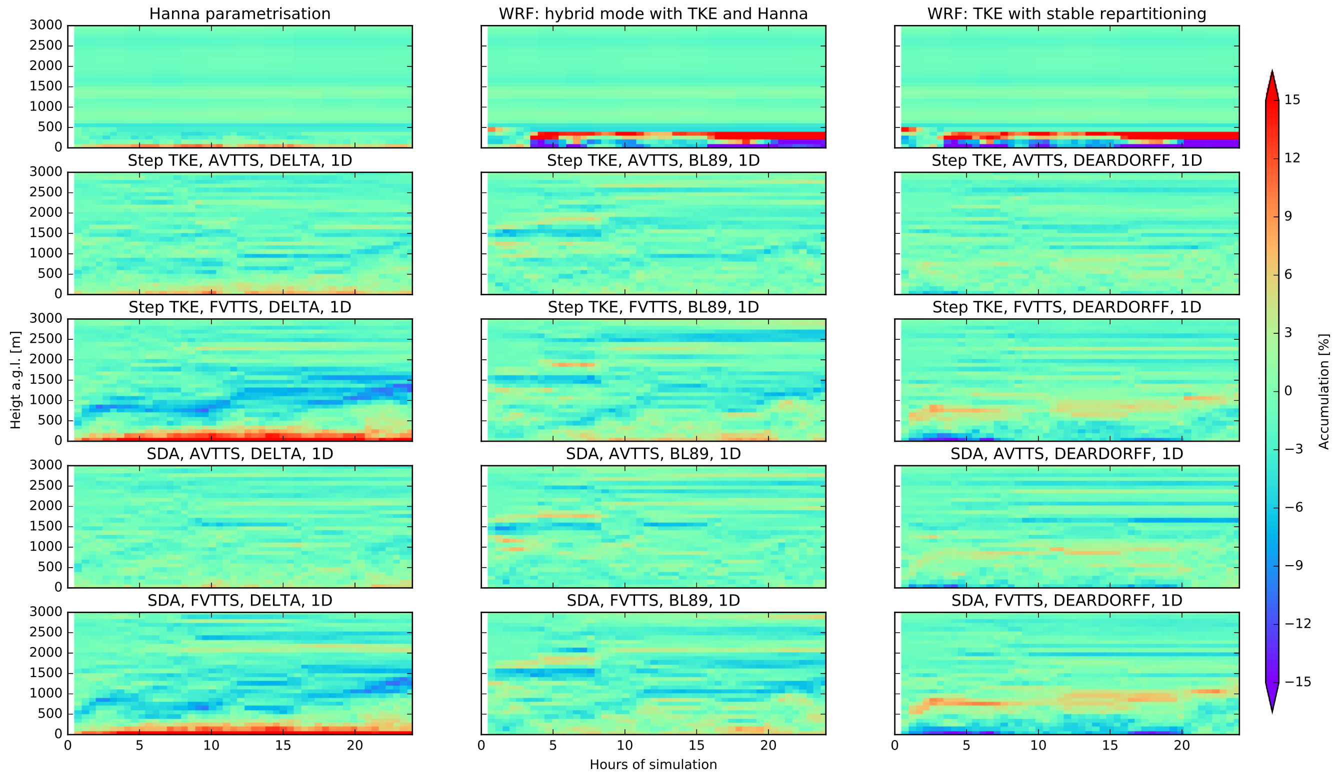


Figure 4. ~~Accumulation~~ The vertical profile of accumulation in well-mixed test ~~in~~ from all the different turbulence configurations in FLEXPART-AROME is shown throughout the 24 hour simulation test. These tests were run in a single column over the ocean surface.

FLEXPART-WRF mode are slightly better with a maximum dilution of 43.3% near the surface and an accumulation of 31.5% at the PBL top.

The ~~bottom-up~~ AVTTS configurations perform consistently better than their ~~top-down~~ FVTTS counterparts. The ~~top-down~~ FVTTS result with DELTA mixing length has the largest surface accumulation of novel FLEXPART-AROME modes (surface accumulation up to 25.7%). The ~~bottom-up~~ AVTTS DEARDORFF mode in a step TKE configuration has the least accumulation and dilution of all models (4.3% and 7.4% respectively), however, use of DEARDORFF is not recommended since it is only valid in a stably stratified atmosphere. ~~Modes TURB_OPTION=11 and 111~~ Aside from the DEARDORFF configuration, modes combining AVTTS with BL89 best conserve the well-mixed state of the passive tracer. ~~With the step TKE performing slightly better~~ The step TKE option performs slightly better than the SDA in this example (0.9% less dilution and 2% more accumulation)~~but in tests over land the~~. Tests over land however showed that SDA had better results. (Appendix C)

The remaining accumulation is due to gradients in mixing length. The DELTA mode has smaller L_w near the surface while DEARDORFF has larger mixing lengths at the surface compared to higher altitudes. We see that mass accumulates in these small mixing length regions.

3.2 Vertical dispersion of a passive surface tracer in the planetary boundary layer

The vertical dispersion of a passive surface tracer is an important test to assure efficient vertical turbulent mixing. The conservation of well-mixedness might be due to inefficient mixing and so, the surface tracer is a necessary supplementary test. We expect the tracer to be well-mixed throughout the turbulent regions within three hours after the initial release.

A point release at the surface at t=0 in a FLEXPART-AROME simulation with isolated vertical turbulent motions for different turbulence modes is shown in fig 5. Figure 5. The final mixing ratio profiles of are shown in appendix D.

Concentrations in FLEXPART-WRF turbulent options are larger compared to new modes. Due to shallow convective mixing in new turbulent modes, particles are allowed to breach the PBL top . The tracer is mixed over a larger vertical range causing further dilution not present in In the Hanna mode and the FLEXPART-WRF modes, the tracer is mixed up to 500m above ground level within the first 3 hours. This corresponds to the maximum boundary layer top within this period. It is obvious however that the tracer is not well mixed in the FLEXPART-WRF turbulent modesconfigurations based on the turbulent kinetic energy.

Similar to the traditional configurations, the novel FLEXPART-AROME turbulent modes succeed in well-mixing the surface tracer within the first three hours. But rather than mixing up to the 500 m above ground level, where the boundary layer top is situated, the novel modes mix the tracer up to an altitude of 1000 m above ground level. This corresponds to the maximum height of the turbulent layer according to the TKE fields in the same period. There is also limited mixing between turbulent en and non-turbulent regions above the shallow convective zone present in the new modes. This in contrast to the sharp PBL in FLEXPART-WRF where all particles are reflected at the PBL top in the isolated turbulence configuration. Note that the use of dynamic TKE fields result in the shifting in time of the convective zone. Particles can be mixed higher up at certain times after which they will no longer mix down but rather remain at the same position.

Due to the inclusion of shallow convective mixing in new turbulent modes, particles are allowed to breach the PBL top and near-surface concentrations in the traditional turbulent option is approximately three times larger compared to the new modes. The tracer is mixed over a larger vertical range causing a dilution not present in Hanna or FLEXPART-WRF turbulent modes. We highlight that, in this case, more than half of the total mass emitted at the surface is transported above the boundary layer by the new turbulent modes. This enables transport along the stronger free tropospheric winds, creating further inconsistencies in dispersion between the traditional and novel turbulent methods.

4 Performance

4.1 Marine Boundary layer tracer

FLEXPART-AROME was built to simulate particle atmospheric transport around Reunion Island to analyse measurements at the high altitude Maïdo observatory Maïdo observatory (Baray et al., 2013). To study the marine boundary layer (MBL) impact on measurements taken at the observatory, we continuously release a passive tracer between 0 and 5 meters above the sea with a lifetime of 24 hours. Results shown are after a spin-up time of 24 hours, LSYNC is set to 300, IFINE and CTL equal 5.

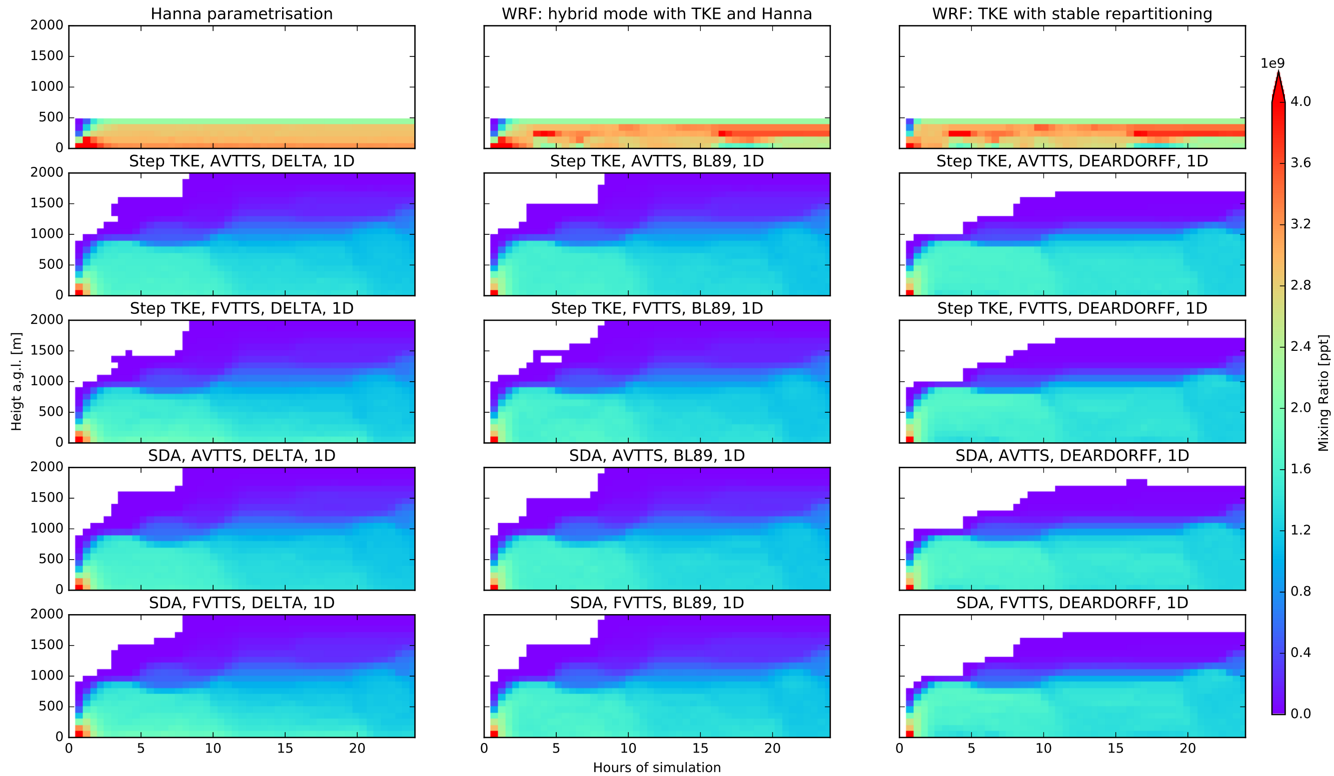


Figure 5. Vertical dispersion of point release at the surface are shown by the time evolution of the vertical mixing ratio profiles throughout the 24 hour simulation test for the different turbulent modes in FLEXPART-AROME. These tests were performed in a single column over the ocean surface.

Due to the strong coupling of the sea-breeze and up-slope mountainous transport the observatory is located in the MBL during the day while at night the reverse process flushes marine tracers with free-tropospheric air as found in isotopic analysis of water vapor at the Maido Maïdo observatory by Guilpart et al. (2017). Figure 6 shows the MBL tracer at Maido usingMaïdo using; i) no turbulent motions, ii) Hanna turbulence and iii) the selected new mode , (TURB_OPTION=0, 1 and 111 respectively). Differences between modes with turbulence are limited in this example. The passive tracer arrives an hour earlier and has a larger vertical distribution when arriving at the observatory in the new mode compared to the performance of Hanna turbulence.

Figure 7 shows the marine boundary layer tracer above a random grid cell at sea. In this figure we clearly see the influence of clouds on the dispersion of passive marine tracer in the vertical. Tracers are convected through strong shallow convection in turbulent clouds that are not resolved in the traditional FLEXPART configuration. Surface mixing ratios in the Hanna mode are elevated compared to those obtained with the new turbulent mode as seen in the point release test.

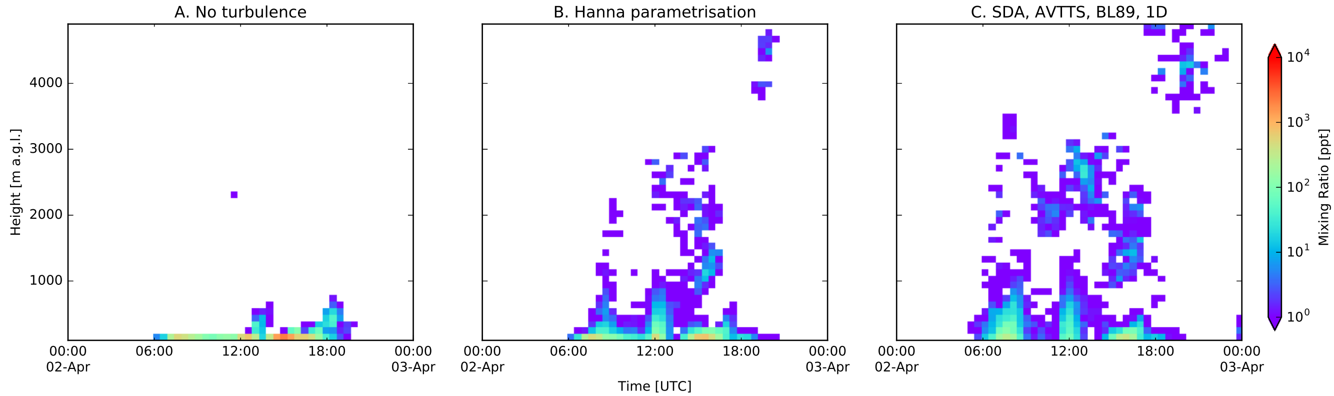


Figure 6. Marine boundary layer tracer profile evolution at the Maïdo observatory. ~~During the day we expect to observe the marine tracer due to efficient coupling of~~ On the ~~sea-breeze and up-slope transport while at night~~ left a simulation with not turbulent motions taken into account. The middle panel shows the ~~observatory is located in air masses of free tropospheric origin~~traditional FLEXPART turbulent mode. ~~Although surface mixing ratios can differ between modes, all of~~ On the ~~configurations produces~~ right hand side we show the ~~desired diurnal cycle at~~ results with the ~~observatory on this particular date~~new turbulent mode.

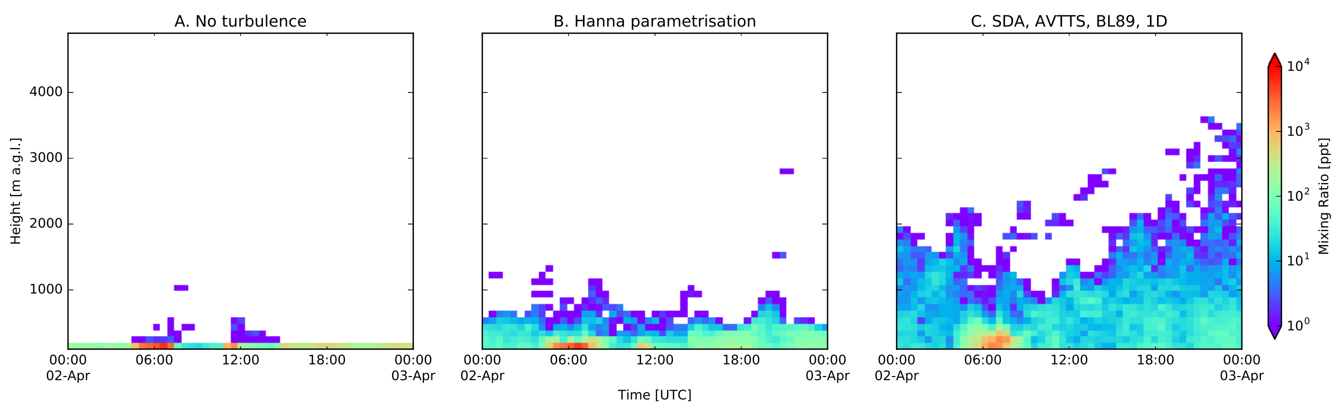


Figure 7. Marine boundary layer tracer profile evolution above sea. ~~We can see that~~ On the ~~new~~ left a simulation with not turbulent ~~mode mixes the passive tracer up toward higher altitudes due to~~ motions taken into account. The middle panel shows the traditional FLEXPART turbulent ~~coupling of shallow convection and clouds~~mode. ~~This behavious is not present in~~ On the ~~original parametrisation and we only have a shallow boundary layer in which~~ right hand side we ~~observe~~ show the ~~marine tracer~~results with the new turbulent mode.

Table 2. Computation time ratios relative to the original Hanna ~~parametrisation~~ parameterization computation time.

Turbulent configuration			TURB_OPTION	Well-mixed test	Point release test	Marine boundary layer run
No turbulent motion			0	0.96	2.30	1.89
Hanna parameterization			1	1.00	1.00	1.00
WRF: hybrid mode with TKE and Hanna			2	0.94	1.18	x
WRF: TKE with stable repartitioning			3	1.12	1.32	x
Step TKE	AVTTS	DELTA	10	4.95	1.06	x
Step TKE	AVTTS	BL89	11	4.89	1.06	x
Step TKE	AVTTS	DEARDORFF	12	6.81	1.06	x
Step TKE	FVTTS	DELTA	20	4.95	1.04	x
Step TKE	FVTTS	BL89	21	4.99	1.05	x
Step TKE	FVTTS	DEARDORFF	22	6.44	1.02	x
SDA	AVTTS	DELTA	110	4.95	1.19	x
SDA	AVTTS	BL89	111	5.21	1.32	1.37
SDA	AVTTS	DEARDORFF	112	9.05	1.24	x
SDA	FVTTS	DELTA	120	5.20	1.17	x
SDA	FVTTS	BL89	121	5.58	1.31	x
SDA	FVTTS	DEARDORFF	122	8.57	1.16	x

4.2 Computation time

We compared the total computation time between the different simulations ran for this work. Simulations were run on a workstation with a single CPU INTEL CORE I7-7700, 32 Gb of DDR4 SDRAM with a GNU compiler. The machine was dedicated to the FLEXPART-AROME simulations to minimise the impact of parallel processes on the computation times. A complete overview of runtimes in reference to the Hanna parameterization are shown in table 2.

Traditionally particles above the PBL are not considered to be turbulent and get advected in one single LSYNC time step. In the new turbulent modes particles above the PBL top are treated in the same way as those below it. This can imply vertical turbulent loops for particles above PBL if the LSYNC input parameter is large. In the well-mixed tests we use the MDOMAINFILL option and initialise a large amount of particles above the PBL. Due to this the relevant novel modes (excluding DEARDORFF) has a mean runtime of 4.8 times that of Hanna. We exclude DEARDORFF in this comparison since: i) its mixing length has no lower limit except the implicit limit imposed by limiting the minimum time step ~~. These~~ and ii) it's use is discouraged since the mixing length is only valid in very specific cases. The DEARDORFF modes have a runtime of 7.5 times the Hanna runtime in testing the well-mixedness.

When running the point release the relevant new modes are 15% slower than the original mode. In the marine boundary layer, ~~TURB_OPTION 111~~ the turbulent mode combining the SDA, AVTTS, and BL89 options in a 1D configuration ran 37% longer than the Hanna ~~parametrisation~~parameterization. We also remark that no turbulent ~~parametrisation~~ parameterization leads to longer run times in these two tests. This is due to the straightforward implementation of turbulent velocities being set to zero. Time steps in displacing the particle are conserved and since the vertical turbulent dispersion is not represented particles remain in regions with a very low time step. ~~A complete overview of runtimes in reference to the Hanna parametrisation are shown in table 2.~~

5 Conclusions

We developed the new FLEXPART-AROME limited domain model version of FLEXPART based on FLEXPART-WRF v3.1.3. This configuration was ~~originally~~ build to model transport around Reunion Island in the Indian Ocean, a small volcanic island which has a complex orographic structure, but can be used with any AROME domain. To simulate turbulence ~~as close to~~ consistently with the operational meteorological model in the region, we implemented new turbulent modes that ingest 3D TKE fields from the NWP. Due to shallow convection ~~energy~~ being taken into account in determining the ~~2D~~ TKE fields in AROME, FLEXPART-AROME is able to represent ~~shallow convective behaviour in the atmosphere.~~ sub-grid scale shallow convective features. There are three important developments that users should consider when selecting the turbulent option that best suits their needs.

– To better represent the local turbulent state of a particle, an adaptive time step was implemented. This configuration is referred to as the adaptive vertical turbulence time step approach and performs consistently better in conserving the well-mixed state of the atmosphere compared to the traditional configuration.

– Turbulent drift in the model is numerically constrained by using the ~~Thomson interface formalism~~ formalism introduced by Thomson et al. (1997). It consists in reflecting or transmitting particles at discrete turbulent interfaces to conserve the well-mixed state of an initially well-mixed atmosphere. Two possible interpretations of ~~the~~ this formalism have been implemented. One approximates turbulence in the FLEXPART-AROME grid by considering every grid-cell to have uniform turbulence with transport being constrained at the ~~vertical~~ boundaries of the model grid and is referred to as the Step TKE option. The other uses ~~a so called~~ the small discontinuity approximation where the turbulent profile is vertically interpolated and transport is constrained at each displacement. ~~To better represent the local turbulent state of a particle we also implemented an adaptive vertical time step in turbulent particle transport. This configuration~~ The latter is referred to as ~~a bottom-up approach and performs consistently better in conserving the well-mixed state of the atmosphere compared to~~ the SDA option. When users are interested in vertical output grids with high resolution, as in the ~~traditional configuration.~~ AROME grid, we advise to use the SDA option. If not, users can select the Step TKE option with lower values of the IFINE and CTL input parameters to speed up the model.

– Three different mixing length ~~parameterisations~~ parameterizations are implemented: DELTA, BL89 and DEARDORFF. Use of the last ~~parameterisation~~ parameterization is discouraged due to it only being valid in stably stratified atmospheres. Users are encouraged to adapt the choice of mixing length parameterization to be in accordance with the NWP.

New turbulent modes have a computation time that is about 5 times larger compared to the Hanna ~~parameterisation~~ parameterization when a large fraction of the particles are above the PBL. However, simulation of tracers predominantly present in the PBL using a new mode in the ~~AROME-SWIO~~ AROME SWIO domain only take 15% longer than the original configuration.

FLEXPART-AROME will be used to study the arrival of marine boundary layer tracers at ~~Maido~~ Maïdo observatory on Reunion island, and the vertical distribution of marine aerosols above the ocean in comparison with measurements. Ingestion of meteorological fields coming from the Meso-NH mesoscale research model will also be introduced in the future to simulate transport at higher resolutions around La Réunion to help study air mass transport on a case study basis.

Code availability. The FLEXPART-AROME code is openly accesible on FLEXPART.eu

Data availability. Data used for the different tests is available upon request.

Appendix A: ~~Conservations of well-mixedness over land~~Different turbulent modes and their respective input parameters

Table A1 shows the different novel turbulent modes implemented in the FLEXPART-AROME code.

Table A1. Different turbulent options introduced in FLEXPART-AROME and their configuration.

TURB_OPTION		AVTTS		FVTTS	
		1D	3D	1D	3D
Step TKE	DELTA	10	15	20	25
	BL89	11	16	21	26
	DEARDORFF	12	17	22	27
SDA	DELTA	110	115	120	125
	BL89	111	116	121	126
	DEARDORFF	112	117	122	127

Appendix B: Implementation of the turbulent mixing length parameterizations

The importance of turbulent mixing length in the new modes is the closing of the turbulent parameterization. Without this value, we have no information on how far particles can mix and so we would have no information on the turbulent time scale.

There are three different implementation of turbulent mixing length L_w. The 1D DELTA L_w is computed as follows:

$$L_w(\text{DELTA,1D}) = \min(0.4 * h(k), \Delta z(k)), \tag{B1}$$

where $h(k)$ and $\Delta z(k)$ represent the height and the thickenss of th k'th model layer respectively. When simulations are run in the 3D mode we use the following formula:

$$L_w(\text{DELTA,3D}) = \min\left(0.4 * h(k), \sqrt[3]{\Delta x \Delta y \Delta z(k)}\right), \tag{B2}$$

where Δx and Δy represent the horizontal resolutions.

The DEARDORFF parameterization is computed by:

$$L_w(\text{DEARDORFF}) = \begin{cases} \sqrt{\frac{2\text{TKE}\theta_{v,ref}}{g\partial\theta_v/\partial z}}, & \text{if} \quad \partial\theta_v/\partial z > 0, \\ \Delta z(k), & \text{otherwise.} \end{cases} \tag{B3}$$

Here, TKE is the local turbulent kinetic energy, $\theta_{v,ref}$ is the virtual potential temperature of the reference state, $\partial\theta_v/\partial z$ is the vertical gradient of the virtual potential temperature and g is earth's gravitational acceleration constant. In FLEXPART-AROME however, the virtual potential temperature is approximated by the potential temperature, neglecting the humidity effect on the air masses.

The BL89 parameterization computes the distance that an air parcel can travel upward and downwards by using the local turbulent kinetic energy and combines both to compute the turbulent mixing lenght:

$$TKE = \int_{z}^{z+l_{up}} \frac{g}{\theta_{v,ref}}(\theta(z') - \theta(z))dz', \tag{B4}$$

$$TKE = \int_{z-l_{down}}^{z} \frac{g}{\theta_{v,ref}}(\theta(z) - \theta(z'))dz', \tag{B5}$$

$$L_w(\text{BL89}) = \left(\frac{l_{up}^{-2/3} + l_{down}^{-2/3}}{2}\right)^{-3/2}. \tag{B6}$$

These equations are solved on the discrete model layers. As a consequence, the minimal mixing length equals Δz. Similar as in the DEARDORFF parameterization, the virtual potential temperatures are approximated by the potential temperatures. The 1D and 3D parameterizations do not differ for both the DEARDORFF and the BL89 parameterizations.

It is important here to note that the DEARDORFF parameterization is the only parameterizations that does not have a lower limit based on the grid definition. It only falls back on the minima of the other implementations when its value becomes negative. The lower limit is rather a computational remnant which stems from the minimal time step. In equation 3 the dt' has a fixed minimum which means that the turbulent time scale is numerically forced to a specific value. When computing τ_w in equation 2 the σ_w value is fixed by the input which means that when its value is forced by the algorithm, we artificially adapt the turbulent mixing length.

Appendix C: Conservations of well-mixedness over land

Shown in ~~figure~~ Figure C1 is the conservation of well-mixedness over land in the morning when the PBL is growing. We see that the DELTA modes all have some accumulation near the surface, the ~~bottom-up~~ AVTTS SDA mode having the least accumulation, similar to the stable PBL over sea. A surface accumulation over land in Hanna in the bottom layer of maximum 14.5%. Comparing the best performing relevant TURB_OPTION parameters 11 and 111 we see that the accumulation in the step TKE mode near the surface is 2.0% larger with the accumulation occurring at the surface from 10 hours simulation onward.

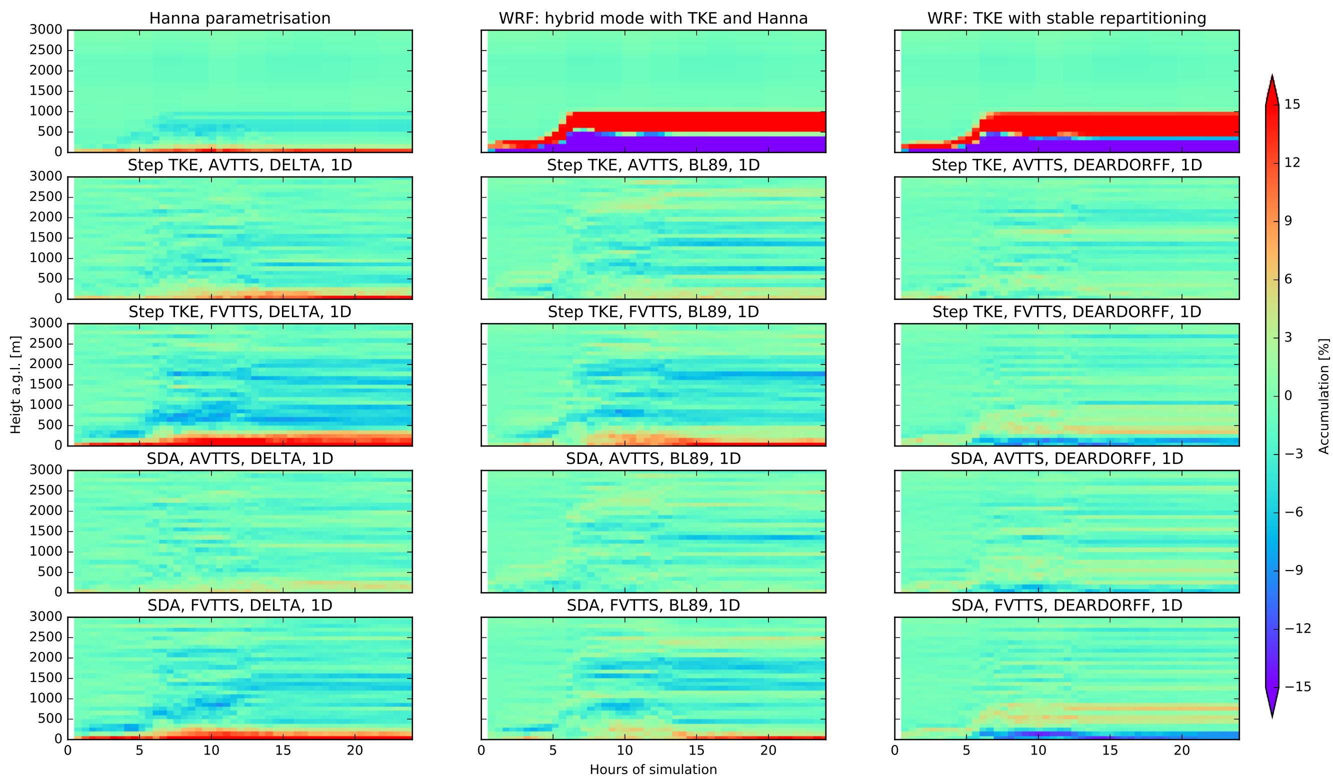


Figure C1. Accumulation in well-mixed test in all different turbulence configurations in FLEXPART-AROME. These tests were run in a column over the ocean surface.

Appendix D: Conservations of well-mixedness over land

After the 24 hour simulation of a passive tracer released at the surface, final mixing ratio profiles for all tested turbulent modes are shown in Figure D1. Due to the shallow PBL in the traditional modes the mixing ratios of the FLEXPART-WRF configurations are a factor 2 to 3 larger. The new turbulent modes are all well mixed near the surface. Due to the shifting convective zone near the top there is no sharp difference between PBL and FT.

We can clearly see two different kinds of mixing between the DEARDORFF parameterizations on the one hand and the DELTA and BL89 modes on the other hand. While DEARDORFF is based on an analytical formula with no real lower limit except the one implicitly imposed by the minimal time step, vertical mixing above the more turbulent layer is slower. This results in a mixing ratio profiles which do not reach as high as the other modes who's lower limit on turbulent mixing length is based on the grid definition.

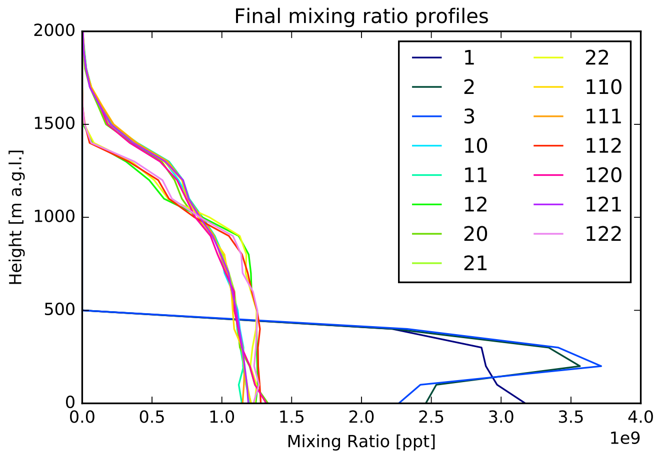


Figure D1. Final mixing ratio profiles in the surface tracers test released over sea. The legend shows the numerical value of the TURB_OPTION parameter input.

Author contributions. Jérome Brioude developped the provisionary FLEXPART-AROME version and adapted FLEXPART-WRF code to ingest AROME data. He supervised and advised Bert Verreyken who was responsible for implementing and testing the Thomson methodology to use 3D TKE fields in the model. Stéphanie Evan was developer on the FLEXPART-WRF version used as a base and a sought after consultant on development of FLEXPART-AROME.

Competing interests. The authors declare that they have no conflict of interest.

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