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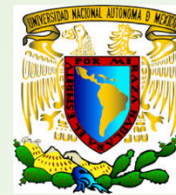
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1 Intercomparison of FLEXPART and CALPUFF dispersion models. An application over a  
2 small tropical island

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### 3 Highlights

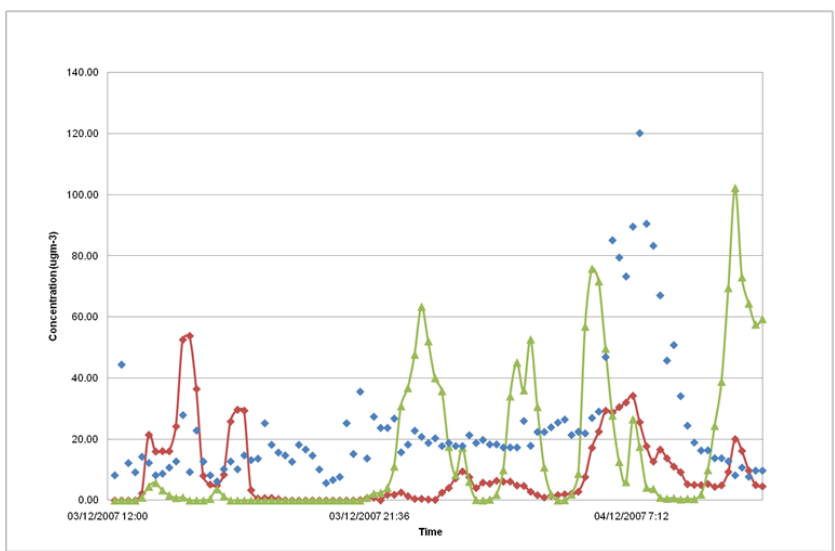
- 4 - FLEXPART/CALPUFF Lagrangian NO<sub>x</sub> dispersion from 60-m smokestack in tropical island
- 5
- 6 - Similar dispersion results using WRF inputs in air quality stations at 2 and 6-km
- 7 - 2-km station FLEXPART results are better than CALPUFF; 6-km results are similar
- 8 - CALPUFF w/o transitional plume rise/stack tip downwash improves its results

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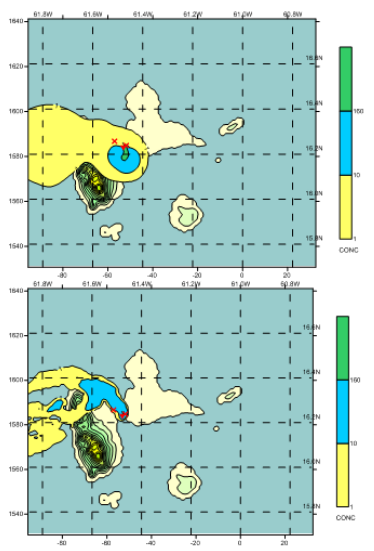
### 11 Graphical Abstract

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## ABSTRACT

A typical practice in air quality modeling assessment is the intercomparison between different dispersion models results and air quality measurements at different atmospheric conditions. In this study, a comparison of the results of two Lagrangian dispersion models, the Lagrangian Particle Dispersion Model FLEXPART and the Lagrangian Puff Model CALPUFF (regulatory model), coupled to the same meteorological fields produced by the Weather Research and Forecasting model (WRF), was done. As a case study, atmospheric dispersion of anthropogenic nitrogen oxides (NO<sub>x</sub>) emissions (considered as a passive tracer) was considered, during a typical case of severe pollution over the densely populated area of the Guadeloupe archipelago (West French Indies), including complex terrain and, of course, coastal influence. Even though Lagrangian models usually provide better results of plume dispersion under strong winds, in this case study weak trade winds are dominant, in order to check both models under non-ideal atmospheric conditions. As a result, compared to NO<sub>x</sub> ground level concentration (glc) observations FLEXPART shows better agreement than CALPUFF. However, as a regulatory model, CALPUFF overestimates both glc observations and FLEXPART maximum NO<sub>x</sub> glc results, with higher values when a higher horizontal resolution is applied. Also, differences between models results arise in the spatial distribution of NO<sub>x</sub> over a 1x1 km<sup>2</sup> horizontal resolution grid domain, showing quite homogenous isopleths with smooth contours for CALPUFF vs. fragmented isopleths with irregular contours for FLEXPART.

**Keywords:** FLEXPART; CALPUFF; Atmospheric dispersion modeling; Tropical island.

### 1. Introduction

Air quality modeling is an essential tool for most air pollution studies (Seinfeld and Pandis, 2012). In that sense, Lagrangian models provide an effective method for simulating atmospheric diffusion when chemical reactions are not relevant. Specifically, Lagrangian models can estimate the local air pollution produced by a large power plant emissions (Souto et al., 2000). They have usually been accepted as the most adequate models for estimating the local plume dispersion from single large point sources (Souto et al., 2009). Currently, the Lagrangian modeling approach is mainly divided into two different categories: particle models and puff models (Zannetti, 2013). In this work, a comparison between two representative Lagrangian models, FLEXPART and CALPUFF, coupled to the WRF meteorological model (Skamarock et al., 2008), is performed.

1 FLEXPART (Stohl et al., 1998) is a widely used Lagrangian particle model that simulates  
2 the transport and dispersion of tracers by calculating the trajectories of a multitude of particles.  
3 As some of the most recent FLEXPART model studies at different regions all over the world,  
4 Wei et al. (2011) used models coupling (WRF-FLEXPART) to describe the impact of high-  
5 pressure conditions on the air quality in northern China. Using the same model coupling, Bei  
6 et al. (2013) analyzed meteorological conditions and plume transport patterns during the Cal-  
7 Mex 2010 study in Tijuana, Mexico. Halse et al. (2013) developed a forecasting system based  
8 on FLEXPART for long-term forecast and assessment of polychlorinated biphenyls  
9 atmospheric transport in different observed air pollution episodes at remote sites in southern  
10 Norway. Also, Arnold et al. (2015) investigated the role of precipitation in the Fukushima  
11 nuclear accident (Tanaka, 2012; Yang, 2014). Besides, Djambazov and Pericleous (2015)  
12 applied the FLEXPART model to estimate how the N air pollutant emissions contribute to the  
13 sea solved nitrogen in the English Channel and the south of the North Sea. Miao et al. (2015)  
14 studied the transport mechanisms of pollutants during a fog event at Bohai Bay, China, using  
15 WRF-FLEXPART models coupling to understand the effects of local atmospheric circulation  
16 and atmospheric boundary layer structure in the air pollution levels. Then, Srinivas et al. (2016)  
17 examined the sensitivity of FLEXPART to the meteorological data inputs simulated by the  
18 mesoscale model ARW in the Kalpakkam coastal environment. Newly, Cheng et al. (2017)  
19 simulated a total monthly CO<sub>2</sub> footprint using FLEXPART.

20 In these studies, FLEXPART has usually been applied to long distance air pollution  
21 transport, with strong circulation patterns providing quite straight trajectories. However,  
22 FLEXPART can also be applied at a local scale in weak wind conditions. As an example, Cécé  
23 et al. (2016) modeled the dispersion of anthropogenic nitrogen oxides (NO<sub>x</sub>) emissions over the  
24 densely populated area of the Guadeloupe archipelago under weak trade winds during a typical  
25 case of severe pollution.

26 CALPUFF (Scire et al., 2000) is one of the most versatile and most applied Lagrangian puff  
27 models used in recent years (Hernández-Garces et al., 2016) which is a multi-layer, multi-  
28 species non-steady-state puff dispersion model, which can simulate the effects of time- and  
29 space-varying meteorological conditions on pollutant transport, transformation, and removal.  
30 CALPUFF contains algorithms for near-source effects such as building downwash, transitional  
31 plume rise, partial plume penetration, subgrid scale terrain interactions as well as longer range  
32 effects such as pollutant removal (wet scavenging and dry deposition), chemical transformation,  
33 vertical wind shear, overwater transport and coastal interaction effects. Also, a regulatory

1 version of the CALPUFF model is available, which tries to guarantee that model results are  
2 mainly higher than the observed air pollution levels.

3 As CALPUFF is a very flexible Lagrangian puff model with different solutions of  
4 atmospheric dispersion phenomena, different validation tests of the CALPUFF model have  
5 been published (O'Neill et al., 2001; Levy et al., 2003; Protonotariou et al., 2004; Cohen et al.,  
6 2005; Yau et al., 2010; Dresser et al., 2011; Fishwick and Scorgie, 2011; Ghannam and El-  
7 Fadel, 2013). As some of the most recent CALPUFF studies, Rood (2014) validated CALPUFF  
8 in an industrial zone in Denver, Colorado, USA. Hernández-Garces et al. (2015a) performed  
9 different CALPUFF assessment by simulating the local dispersion of SO<sub>2</sub> (as a passive tracer)  
10 from a large power plant smokestack (356.5 m height, with four parallel independent pipes  
11 inside the same concrete structure), considering both different stack modeling settings (four  
12 pipes vs. one equivalent single pipe) and meteorological inputs (both meteorological models  
13 results and observations). Pivato (2015) applied CALPUFF to assess the airborne concentration  
14 of an emitted pesticide. Holnicki et al. (2016) presented a case study application of the  
15 CALPUFF model at an urban scale over the Warsaw area. More recently, Fallah-Shorshani et  
16 al. (2017) simulated the pollutants transport at the top of the urban canopy using CALPUFF as  
17 part of an integrated modeling system to simulate ambient nitrogen dioxide in a dense urban  
18 neighborhood.

19 Apart from the mesoscale wind provided as input to any Lagrangian model, two main  
20 phenomena affect these model's performance at the local scale: the atmospheric turbulence, as  
21 pollutant atmospheric diffusion depends on it; and the planetary boundary layer (PBL) depth,  
22 because pollutants plume rise is usually limited by this depth.

23 About the atmospheric turbulence effect, both FLEXPART and CALPUFF models follow  
24 the classical Lagrangian approach of increasing the element (either puff or particle) position  
25 every time step  $\Delta t$  by adding a  $v\Delta t$  distance. However, in the case of the FLEXPART particle  
26 model,  $v$  includes not only the mesoscale wind speed but also the turbulent wind fluctuations  
27 effect, in order to estimate the pollutants' atmospheric diffusion; this FLEXPART approach is  
28 based on the parameterization scheme for statistical models proposed by Hanna (1982),  
29 modified by Ryall et al. (1997). On the other hand, CALPUFF represents atmospheric diffusion  
30 using a piece-wise Gaussian distribution, as in previous Adaptive Puff Models (Ludwig et al.,  
31 1989; Souto et al., 1998). Although CALPUFF provides several options to estimate atmospheric  
32 Gaussian diffusion parameters, the classical approach, based on the similarity theory (Monin  
33 and Obukhov, 1954), was adopted.

1 About PBL depth, the applied estimation with both models is based on the bulk Richardson  
2 approach of Vogelezang and Holtslag (1996) with different convective velocity scale,  $w^*$ .

3 CALPUFF coupled to WRF meteorological model (Skamarock et al., 2008) derives this  
4 friction velocity from the vertical eddy fluxes of momentum WRF results ( $|w'u'|$ ), using the  
5 MMIF meteorological preprocessor (Brashers and Emery, 2016) that applies Deardorff (1970)  
6 expression,

$$7 \quad w_* = \sqrt[3]{\frac{(\overline{w'\theta'_v})gz_i}{T_v}} \quad (1)$$

8 where  $g$  is gravitational acceleration,  $T_v$  is absolute temperature,  $z_i$  is the average depth of the  
9 mixed layer, and  $\overline{w'\theta'_v}_0$  is the kinematic vertical turbulent flux of virtual potential  
10 temperature near the surface (Stull, 1988).

11 On the other hand, FLEXPART follows the convective velocity scale suggested by  
12 Vogelezang and Holtslag (1996),

$$13 \quad w_* = \sqrt[3]{\frac{(\overline{w'\theta'_v})_0gh_{mix}}{\theta'_{v1}}} \quad (2)$$

14 where  $\overline{w'\theta'_v}_0$  is the heat flux,  $g$  is the acceleration due to gravity,  $h_{mix}$  is the PBL height, and  
15  $\theta'_{v1}$  is the improved temperature scale at the 1<sup>st</sup> model level (Holtslag and Nieuwstadt, 1986).

16 Both  $w^*$  formulations are analogous, thus similar PBL depth estimations are expected for  
17 simulations obtained with CALPUFF and FLEXPART. However, MMIF also incorporates the  
18 methodology of Gryning and Batchvarova (2003), which reduces the critical Richardson  
19 number for overwater PBL heights from 0.25 to 0.05. Therefore, in a small island domain, it is  
20 expected that overwater MMIF PBL depth will be smaller than the FLEXPART PBL depth  
21 estimation.

22 This is just one example of the influence of the simulation domain conditions in model  
23 assessment and intercomparison. Therefore, it is common to compare models results at different  
24 conditions. Different CALPUFF and AERMOD regulatory model comparisons were published  
25 (Dresser et al., 2011; Tartakovsky et al., 2013; Gulia et al., 2015; Tartakovsky et al., 2016).  
26 Also, Scire et al. (2013) added a comparison with CAMx (Tesche et al., 2001) using its Plume-  
27 in-Grid approach. Rood (2014) performed a comparison between former regulatory model  
28 Industrial Source Complex 2 (US-EPA, 1992) and RATCHET (Ramsdell et al., 1994). Other  
29 researchers compared CALPUFF with different models: Yau et al. (2010) against  
30 AUSTAL2000 (VDI, 2000); Protonotariou et al. (2004) against Eulerian models UAM (SAI,  
31 1992) and REMSAD (ICF Consulting, 2002); Chang et al. (2003) against HPAC (DTRA, 1999)  
32 and VLSTRACK (Bauer and Gibbs, 1998).



1 Also, some FLEXPART model comparisons were performed at long distance experiments.  
2 Probably the most comprehensive is the comparison against HYSPLIT (Draxler and Hess,  
3 1998), CALPUFF and other two dispersion models in the European Tracer Experiment (ETEX)  
4 and the Cross-Appalachian Tracer Experiment (CAPTEX), using a tracer cloud dispersion. This  
5 last study concluded that CALPUFF performance was significantly poorer than the  
6 performance of the other three models in the ETEX experiment, while CALPUFF results were  
7 improved in CAPTEX (Anderson and Brode, 2010). At a local scale, Souto et al. (2001)  
8 compared two Lagrangian models during an SO<sub>2</sub> air pollution episode, the Lagrangian particle  
9 model (LPM, Pielke, 1984) and the Adaptive Puff Model 2 (APM2, Souto et al., 1998),  
10 concluding that, even though vertical concentration profiles provided by LPM are more  
11 heterogeneous than estimated by APM2 (with the same meteorological input), this puff model  
12 achieved a better agreement to g/c observations than LPM, as APM2 was able to reproduce  
13 observed local hotspots, which were not estimated by LPM. However, previous studies at  
14 different domains indicated that Lagrangian particle models usually provide better results than  
15 other simple Gaussian puff models (Saltbones et al., 1996). Therefore, model performance  
16 depends not only on the basic modeling approach (i.e., particles vs. puffs) but also on the  
17 modeling solutions for specific phenomena (plume rise, building effects, ...) and on the domain  
18 conditions (Hernández-Garces et al., 2015b).

19 This is the reason why in this work a singular simulation domain was selected, the  
20 Guadeloupe archipelago, including the complexity of small orographic tropical islands  
21 (width < 50 km) with strong sea influence at the Caribbean Region. Particularly, from medium  
22 to weak trade winds (<7 ms<sup>-1</sup>), its complex terrain at the Lesser Antilles may induce thermal  
23 and orographic local circulations, which strongly affect the air quality levels (Cécé et al 2014,  
24 Cécé et al 2016).

25 In this study, a comparison of the results of two Lagrangian dispersion models, FLEXPART  
26 and CALPUFF, coupled to the same meteorological fields input provided by WRF model  
27 (Skamarock et al., 2008), was done, considering NO<sub>x</sub> emissions as a passive tracer over the  
28 densely populated area of the Guadeloupe archipelago under weak trade winds, during a typical  
29 episode of severe pollution.

30

## 31 **2. Materials and methods**

32

### 33 *2.1. Study location*

1 The Guadeloupe archipelago is located in the middle of the Lesser Antilles arc at 16.22° N  
2 and 61.55° W (Figure 1). This archipelago includes two main islands: Basse-Terre, a complex  
3 terrain island with a maximum elevation height of 1 467 asl-m, and Grande-Terre, a flat terrain  
4 island with a maximum elevation height of 135 asl-m. The archipelago islands form a basin  
5 between the two main islands. This region is usually characterized by calm winds, but specific  
6 airflows in the atmospheric boundary layer can be observed with strong sea influence along its  
7 narrow coastline.

8 The main anthropogenic sources of atmospheric pollution in this region are a diesel power  
9 plant (PWP, Figure 2) located in the center of the archipelago (Figure 1) and vehicles on the  
10 primary road network. As this is the largest NO<sub>x</sub> emission source in the region, observed NO<sub>x</sub>  
11 glc is mainly produced by PWP emissions, so the NO<sub>x</sub> emission can be considered as a tracer  
12 of its plume. Two air quality stations near to the PWP (1 878 m and 6 135 m travel distances)  
13 are recording data every 15-min, that will be applied in model assessment.

14

15 Here Fig. 1

16

17 Here Fig. 2 Diesel power plant.

18

## 19 *2.2. Simulation period and WRF meteorological results*

20 Cécé et al. (2016) studied a period of 24-hours (1200 LT 3 December-1200 LT 4 December  
21 2007) with high NO<sub>x</sub> glc and weak winds, as the most typical meteorological conditions that  
22 produce poor air quality at the two sites available. To evaluate the performance of the two  
23 selected Lagrangian models, FLEXPART and CALPUFF, the same period was chosen for  
24 model assessment.

25 Meteorological input for both Lagrangian models was obtained from the WRF model  
26 (Skamarock et al., 2008) previously validated simulations (Cécé et al., 2016). Because of the  
27 complex terrain and strong sea influence around the domain, six one-way WRF nested grids  
28 (Figure 3) were applied from 27-km horizontal resolution (D1) to 111-m horizontal resolution  
29 (D6). However, just the innermost grids WRF results (D4, D5, D6) were applied as  
30 meteorological inputs for the air quality simulations.

31 Taking into account the large differences between the six grids applied, the following WRF  
32 model settings were carefully selected to get the best model performance: 70 unequally spaced  
33 half vertical eta-levels with the lowest half level at 13 agl-m and the model top set at 100 hPa  
34 pressure level; Rayleigh damping on vertical velocity with a damping layer depth of 5 km;

1 Monin-Obukhov similarity; WRF single-moment 6-class microphysics scheme; Rapid  
2 Radiative Transfer Model longwave scheme (Mlawer et al., 1997), the Dudhia shortwave  
3 scheme (Dudhia, 1989); and ensemble-mean non-local-K YSU scheme for the PBL in  
4 mesoscale domains (Hong et al., 2006).

5

6 Here Fig. 3

7

8 The topography of the Guadeloupe archipelago (domains D4, D5 and D6) was interpolated  
9 from the Institut Géographique National 50-m topographic map and its land-use was  
10 interpolated from the 25-m Corine Land Cover map (EEA, 2007) pre-converted to 24 USGS  
11 land-use categories (Anderson et al., 1976; Pineda et al., 2004).

12

Here Fig. 4

13 As D4 to D6 grids provide the meteorological inputs for air quality simulations, Figure 4  
14 shows the three wind roses obtained from the corresponding grid WRF output at the PWP  
15 location. As it is observed in the three domains, changes in wind direction are covering mostly  
16 360°, as in a weak wind condition. However, sea breeze regimes are significant, because some  
17 more frequent wind directions are observed, with differences depending on the grid resolution  
18 considered: NNE, E and ESE wind directions are the most frequent using D4 (1 km resolution)  
19 and D5 (333 m resolution) grids outputs; however, D6 (111 m resolution) grid output also shows  
20 frequent W and E wind directions. In addition, wind speed (Figure 5) is quite different  
21 depending on the grid resolution, with frequent values up to 5.0 ms<sup>-1</sup> from D4 output, which are  
22 lower from D6 output.

Here Fig. 5

23 There is more short-term variability in the D5 and D6 wind speeds especially in the daytime.  
24 It is mainly linked with the much finer resolution and the better representation of the lower-  
25 levels daytime convection turbulence.

26 These differences in the WRF meteorological results depending on the grid resolution can  
27 be relevant in air quality simulations in this domain. Therefore, the same meteorological input  
28 will be set for both Lagrangian models simulations.

29

### 1 2.3. FLEXPART model setup

2 Because of the domain complexity and strong meteorological variability along with the 24-  
3 h simulation, 10-min. WRF outputs are provided to the FLEXPART model, following Cécé et  
4 al. (2016). These meteorological outputs include mass-weighted time-averaged wind fields,  
5 friction velocity, heat sensible flux and PBL height. PBL turbulence is parameterized following  
6 the Hanna scheme (Hanna, 1982) that computes turbulent profiles depending on the  
7 atmospheric stability of the PBL.

8 According to the Pollutant Emissions French Register, PWP released 9.79 kilotons of NO<sub>x</sub>  
9 (equivalent NO<sub>2</sub>) in 2007. Based on this annual amount, NO<sub>x</sub> total mass emitted by PWP during  
10 24 h is set to 26.82 tons, with a constant emission rate, as no hourly profile information is  
11 available. The PWP plume is represented as Cécé et al. (2016) did using plume observational  
12 pictures (Figure 6) by a volume of 30x30x340 m centered at 61.5515°W and 16.2280°N. The  
13 plume base corresponds with a smokestack height of 60 m AGL.

14

15 Here Fig. 6

16

17 In the FLEXPART model, the concentrations are calculated on the basis of the number of  
18 trajectories located within each grid cell and their mass fraction. The larger the number of  
19 released particles is, the better the particle mass fraction statistics for each grid cell are  
20 represented. Hence, for a given resolution grid, an optimal number of trajectories to run in the  
21 model needs to be estimated. In order to determine this number corresponding to result stability,  
22 several simulation tests have been made with a number of particles released every 15 min  
23 ranging from 5000 to 60,000 particles. These tests have shown that the result stability was  
24 reached for 1-km grid, 333-m grid and 111-m grid, with the respective number of particles  
25 emitted every 15 min during the 24-h period: 20,000, 30,000 and 50,000 (Cécé et al., 2016).

26 NO<sub>x</sub> dispersion emitted from the PWP is simulated over domains D4, D5 and D6  
27 (simulations: flx1km, flx0.3km, flx0.1km). As a result, NO<sub>x</sub> concentrations are computed every  
28 15 min. at the two available air quality stations, AQS1 (Pointe-à-Pitre) and AQS2 (Baie-  
29 Mahault). NO<sub>x</sub> concentrations are also computed over a three-dimensional grid, with 38 terrain-  
30 following levels (first level at 10 agl-m and top level at 3 000 agl-m) and 1 km, 0.3 km and 0.1  
31 km of horizontal resolution, respectively. However, following PWP plume observational  
32 pictures (Cécé et al., 2016), a maximum plume top is set at 400 agl-m.

33

1 *2.4. CALPUFF model setup*

2 CALPUFF model setting follows default values except for the critical Gaussian dispersion  
3 coefficients,  $\sigma_y$  and  $\sigma_z$ , which are dynamically calculated from  $\sigma_v$  and  $\sigma_w$  micrometeorological  
4 variables values provided by WRF outputs. PWP emission source representation applied in  
5 FLEXPART model was adapted to CALPUFF model using the same size. Also, the same WRF  
6 meteorological outputs were applied to obtain three different air quality simulations  
7 (simulations: puff1km, puff0.3km, puff0.1km). However, MMIF preprocessor (Brashers and  
8 Emery, 2016) was required to adapt WRF outputs are CALPUFF meteorological inputs; in this  
9 process, new PBL depth values were computed by MMIF with a vertical resolution 20 times  
10 the WRF vertical resolution.

11 Similar to FLEXPART, 37 vertical layers to calculate NO<sub>x</sub> concentrations were set, although  
12 with the lowest level at 20 m because of CALPUFF constraints.

13 To compare CALPUFF in conditions as close as possible to FLEXPART a second simplified  
14 CALPUFF setting was tested (simulations: puff1kmS, puff0.3kmS, puff0.1kmS), considering  
15 neither the transitional plume rise nor the stack tip downwash, and setting a uniform vertical  
16 distribution in the near field; as these local phenomena are not solved by FLEXPART.

17 Therefore, considering this simplified CALPUFF setting, just two main differences between  
18 FLEXPART and CALPUFF settings remain: the lowest vertical level height and the PBL  
19 scheme used. Table I summarizes the different configurations applied, classified into two  
20 groups.

21

22 Here **Table I**

23

24 *2.5 Model assessment and intercomparison*

25 To obtain a quantitative assessment of FLEXPART and CALPUFF simulations the BOOT  
26 Statistical Model Evaluation Software Package, version 2.01, as distributed with the Model  
27 Validation Kit (Chang and Hanna, 2005) was applied. In this study, BOOT package was applied  
28 to compare 15-min estimated and observed ground level concentrations. Four statistics were  
29 considered,

30 Fractional bias (FB),

31 
$$FB = \frac{(\overline{c_o} - \overline{c_p})}{0.5(\overline{c_o} + \overline{c_p})} \quad (3)$$

32 Underpredicting component of the FB (FBFN),

$$FBFN = \frac{\frac{1}{2}\sum_i[|C_{oi}-C_{pi}|+(C_{oi}-C_{pi})]}{\frac{1}{2}\sum_i(C_{oi}+C_{pi})} \quad (4)$$

Overpredicting component of the FB (FBFP),

$$FBFP = \frac{\frac{1}{2}\sum_i[|C_{oi}-C_{pi}|+(C_{pi}-C_{oi})]}{\frac{1}{2}\sum_i(C_{oi}+C_{pi})} \quad (5)$$

Normalized mean square error (NMSE),

$$NMSE = \frac{\overline{(C_o - C_p)^2}}{\overline{C_o C_p}} \quad (6)$$

where  $C_p$  denotes model predictions,  $C_o$  denotes observations,  $C_{oi}$  is the  $i^{\text{th}}$  observed value,  $C_{pi}$  is the  $i^{\text{th}}$  predicted value and overbar ( $\overline{C}$ ) denotes the average over the dataset.

This quantitative assessment is complemented by the graphical comparison of ground level concentration time series from model results and observations at the two air quality sites.

Because of the limited air quality monitoring sites available in the region (Figure 1), to intercompare the models' skills, two different approaches were applied: (a) qualitative comparison of simulated spatial plumes transport, and, (b) maximum plume impact comparison (Souto et al., 2014), including,

- the simulated maximum  $\text{NO}_x$  glc time series,  $C_{\text{max}}$ , over the simulation grid,
- the travel distance to the maximum glc time series,  $X_{\text{max}}$ , and,
- the spatial distribution of all the maximum glc locations along the simulation period.

These intercomparison features can be useful to explain models' results differences and, also, previous model assessment.

### 3. Results

#### 3.1 Graphical comparison between simulated and observed time series of $\text{NO}_x$ concentrations.

To compare the results of two Lagrangian dispersion models, FLEXPART and CALPUFF, against observations, FLEXPART (flx1km, flx0.3km, flx0.1km), CALPUFF (puff1km, puff0.3km, puff0.1km) simulations results and observed 15-min  $\text{NO}_x$  ground level concentration-time series at two stations are shown in Figure 7.

Here Fig. 7

In all simulations both models have similar tracer arrival time at both stations, that is,  $\text{NO}_x$  glc first significant values started around 0 UTC 4 December at AQS1, and around 12 UTC 3 December at AQS2; although, from the observations time series,  $\text{NO}_x$  seems to arrive an earlier at AQS1, around 22 UTC 3 December.

1 Considering the observed  $\text{NO}_x$  glc peaks, the largest one in AQS2 ( $120 \mu\text{g m}^{-3}$ ) is observed  
2 at 07 UTC 4 December: all the simulations reproduce glc peaks around that time, but with lower  
3 values. This modeling peak underestimation occurs near primary road network and at rush-hour  
4 traffic, so observed peak seems linked with vehicle  $\text{NO}_x$  emissions not taken into account in the  
5 models. The largest peaks (below  $80 \mu\text{g m}^{-3}$ ) is obtained in flx1km simulation, but 2 h before  
6 the observed peak, and much lower peaks (below  $60 \mu\text{g m}^{-3}$ ) in time from flx0.3km, flx0.1km  
7 simulations; also puff1km simulation achieves a significant peak in time, but lower (below  $40$   
8  $\mu\text{g m}^{-3}$ ) than others simulations. In fact, both models get higher peaks several hours before the  
9 observed peaks, and these modelled peaks are not always at the same time for both models:  
10 even though it is well known the high sensitivity of plume transport to any errors in the wind  
11 direction provided by WRF model, also the turbulent diffusion approaches applied for each  
12 model have a significant influence in these results.

13 As an example, in Figure 7a CALPUFF simulation obtains a significant glc peak at AQS1  
14 at 4:15 UTC 4 December; however, glc observations do not show any peak, in agreement with  
15 FLEXPART glc results. Otherwise, FLEXPART reproduces, in good agreement, the highest  
16 peak ( $200 \mu\text{g m}^{-3}$ ) observed in AQS1 at 5 UTC 4 December, while all the CALPUFF  
17 simulations overestimate (close to  $1\ 000 \mu\text{g m}^{-3}$ ) that observed peak.

18

### 19 *3.2 Statistical evaluation results of $\text{NO}_x$ simulated concentrations*

20 From the 15-min glc time series comparison some significant differences between observed  
21 and modeled ground-level concentrations arise. Therefore, statistics provide a quantitative  
22 estimation of model errors.

23 Table II shows the BOOT package statistical results obtained from FLEXPART and  
24 CALPUFF simulations results against observations at AQS1 and AQS2.

25

26 Here **Table II**

27

28 From the core statistics, FB and RMSE, better results were obtained using FLEXPART vs.  
29 CALPUFF, especially at AQS1, mainly because of the large CALPUFF glc peaks  
30 overestimations. Moreover, the higher meteorological resolution does not guarantee better  
31 CALPUFF results, with the worst results in puff0.3km at AQS1. However, using the  
32 FLEXPART flx0.1km simulation achieves the best statistics at AQS1.

1 On the other hand, CALPUFF statistics are significantly better at AQS2 vs. AQS1, while  
2 FLEXPART statistics are similar at both stations. Also, the increase of the meteorological  
3 resolution at AQS2 improves the FLEXPART results, with the minima NMSE and FB values  
4 from flx0.1km simulation. This effect is not always observed in CALPUFF results.

5 As expected, FBFP values show a pronounced overprediction of the observed concentrations  
6 at AQS1 in CALPUFF simulations, due to their too large modeled peaks. This FBFP  
7 overprediction of FB significantly exceeds the underprediction component FBFN. At the  
8 opposite, at AQS2 FBFN exceeds FBFP for both models simulations, that is, both models  
9 underestimate the observed concentrations along the simulation period; showing that models  
10 results are extremely sensitive not only to the meteorological conditions (which are the quite  
11 similar at both stations) but also to the station relative location from the emission source.  
12 Therefore, in the following sections other model comparisons and assessment approaches are  
13 also applied.

### 14 15 *3.3 Spatial distribution of NO<sub>x</sub>*

16 Considering previous statistics, FLEXPART model performance using three different  
17 horizontal resolutions (1 km, 333 m, and 111 m) is better as the meteorological input grid  
18 resolution is finer. Therefore, as the clearest NO<sub>x</sub> glc spatial distribution, Figure 8 shows the  
19 models glc results over the 1 km horizontal resolution domain (D4). Compact isopleths with  
20 smooth contours from CALPUFF (Fig. 8a) and fragmented isopleths with irregular contours  
21 from FLEXPART (Fig. 8b) are observed. Particularly, the influence of two topographic tops at  
22 the west of the source is apparent in the fragmented FLEXPART glc distribution; not observed  
23 in the compact CALPUFF glc distribution.

24  
25 Here Fig. 8

### 26 27 *3.4 Plume impact evaluation of simulations*

28 Plume impact evaluation based on the simulated maximum NO<sub>x</sub> glc, C<sub>max</sub>, over the  
29 simulation grid is shown in Figure 9, both for FLEXPART and CALPUFF at the three different  
30 grids resolutions. Unfortunately, it is not possible to estimate C<sub>max</sub> from glc observations for  
31 model assessment, because only two stations data are available (Souto et al., 2014).

32  
33 Here Fig. 9



1 For NO<sub>x</sub> maximum glc, CALPUFF results are significantly higher than FLEXPART results,  
2 and this difference increases with higher horizontal resolutions. These comparative results are  
3 consistent with the CALPUFF regulatory model condition, that guarantees glc overestimation  
4 in any condition. However, so large differences suggest that FLEXPART can be a better option  
5 for more accurate regulatory purpose in this complex domain if its systematic validation is  
6 performed.

7 Following wind direction independent travel distance to maximum concentration location,  
8 X<sub>max</sub> (Figure 10), FLEXPART estimates higher maximum distance values than CALPUFF,  
9 while minimum values are very similar. As maximum X<sub>max</sub> values usually correspond to  
10 unstable conditions, with higher PBL depth, these comparative results should be a consequence  
11 of the different PBL depth schemes applied by each model in unstable conditions. Also, higher  
12 X<sub>max</sub> values should decrease the estimated NO<sub>x</sub> glc, both maximum value, C<sub>max</sub>, and glc  
13 values at the two air quality stations. Because these stations travel distances are lower than  
14 FLEXPART X<sub>max</sub> values but similar to CALPUFF X<sub>max</sub> values. These differences can be  
15 related to the glc overestimation observed in CALPUFF statistical assessment.

16 Besides, X<sub>max</sub> maximum values are higher with lower horizontal resolutions, both in  
17 FLEXPART and CALPUFF simulations; also with differences between X<sub>max</sub> fluctuations  
18 along the time, again depending on the resolution.

19

20 Here Fig. 10

21

### 22 *3.5 Spatial distribution of location of simulated maximum glc*

23 The spatial distributions of estimated maximum glc locations using CALPUFF and  
24 FLEXPART models are shown in Figure 11. CALPUFF simulations at three different  
25 horizontal resolutions show a reduced maximum impact area, close the emission source, and  
26 close to the air quality stations. This feature can produce high glc estimations at station  
27 locations, that justify the overestimation observed in CALPUFF statistical assessment.

28 At the opposite, FLEXPART simulations produce different maximum impact areas than  
29 CALPUFF simulations using the same horizontal resolution. Also, FLEXPART maximum  
30 impact areas are different using different resolutions, as this model seems to be sensitive to the  
31 meteorological input horizontal resolution.

1 Here Fig. 11

### 2 *3.6 Simplified CALPUFF setup*

3 Statistical assessment of default CALPUFF setup, including original schemes for some  
4 features, as the transitional plume rise and the stack tip downwash, shows worse results than  
5 FLEXPART model results. However, for a better intercomparison between both models  
6 excluding these CALPUFF original schemes (not considered in the FLEXPART model), a  
7 second simplified CALPUFF setup is evaluated.

8 Table III shows the statistical results from BOOT software obtained from this second  
9 simplified CALPUFF setup simulations, compared to the observations at AQS1 and AQS2  
10 stations.

11

12 Here **Table III**

13

14 Although statistics show a poor CALPUFF performance, comparing with CALPUFF default  
15 setup simulations assessment (Table II) a significant improvement is observed. Therefore,  
16 original CALPUFF schemes for plume rise and stack tip downwash are not recommended in  
17 this case study. Otherwise, CALPUFF performance is still far from the FLEXPART statistics,  
18 as significant differences in model's results remains.

19

## 20 **4. Conclusions**

21 Two Lagrangian atmospheric dispersion approaches, particle and puff models are usually  
22 applied to estimate passive pollutants dispersion over a complex terrain environment. In this  
23 work, a singular simulation domain, the Guadeloupe archipelago was selected to test the  
24 FLEXPART Lagrangian particle model and CALPUFF Lagrangian puff model. This domain  
25 includes several islands with complex terrain and strong sea influence in the Caribbean Region.  
26 Also, weak winds conditions were selected, as they produce the highest air pollution levels  
27 observed in this region. NO<sub>x</sub> emissions from a single power plant located on the main island  
28 were considered as a passive tracer.

29 Both Lagrangian models were coupled to the same WRF meteorological results at three  
30 different high-resolution grids (up to 111 m), to test their relative accuracy by comparison to  
31 15-min ground level concentration observations at two air quality stations close to the emission  
32 source. During the testing period, the statistical model assessment shows that FLEXPART  
33 achieves significantly better agreement with glc observations than CALPUFF.

1 About the models' intercomparison, the spatial distribution of NO<sub>x</sub> shows homogenous  
2 isopleths with smooth contours for CALPUFF, while fragmented isopleths with irregular  
3 contours are observed with FLEXPART, showing the effect of the complex topography over  
4 the NO<sub>x</sub> plume. Considering the estimated maximum NO<sub>x</sub> g/c, CALPUFF results are much  
5 bigger than FLEXPART, and CALPUFF results increase with the meteorological input  
6 resolution. Travel distance to the maximum g/c is usually longer with FLEXPART than with  
7 CALPUFF, and further away from the air quality stations. This feature has in agreement with  
8 the higher CALPUFF maximum g/c results and, also, its g/c overestimation observed in this  
9 model assessment. Also, maximum g/c locations from CALPUFF results are concentrated in a  
10 small area close to the air quality stations; while the corresponding FLEXPART locations are  
11 further away from the emission source, and they extend over larger areas.

12

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18

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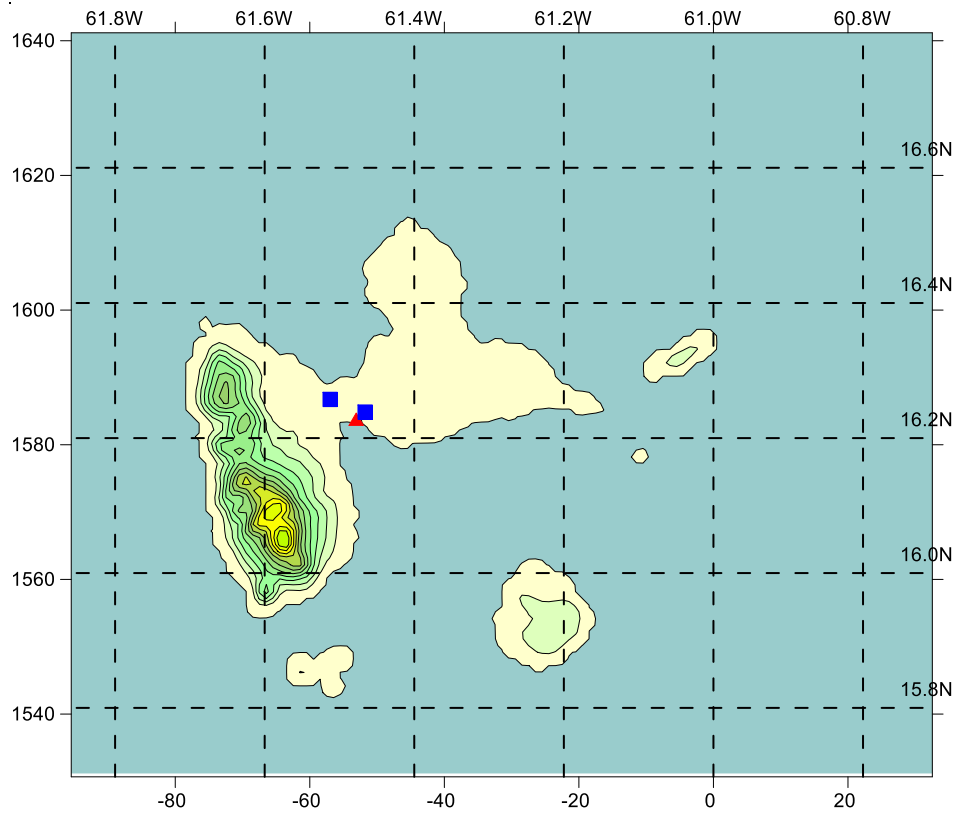
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1

2 Fig. 1 Map of the Guadeloupe archipelago, with the main NO<sub>x</sub> emission source, PWP (red  
3 triangle), and two air quality stations near it (blue squares): AQS1 (Pointe-à-Pitre, 1 878 m  
4 travel distance) and AQS2 (Baie-Mahault, 6 135 m travel distance).

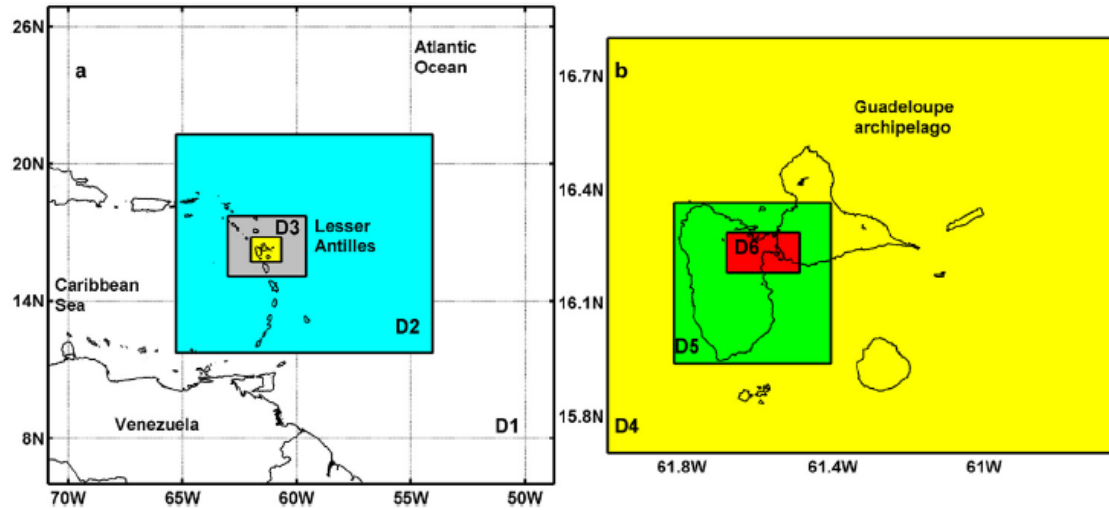
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2 Fig. 2 Diesel power plant

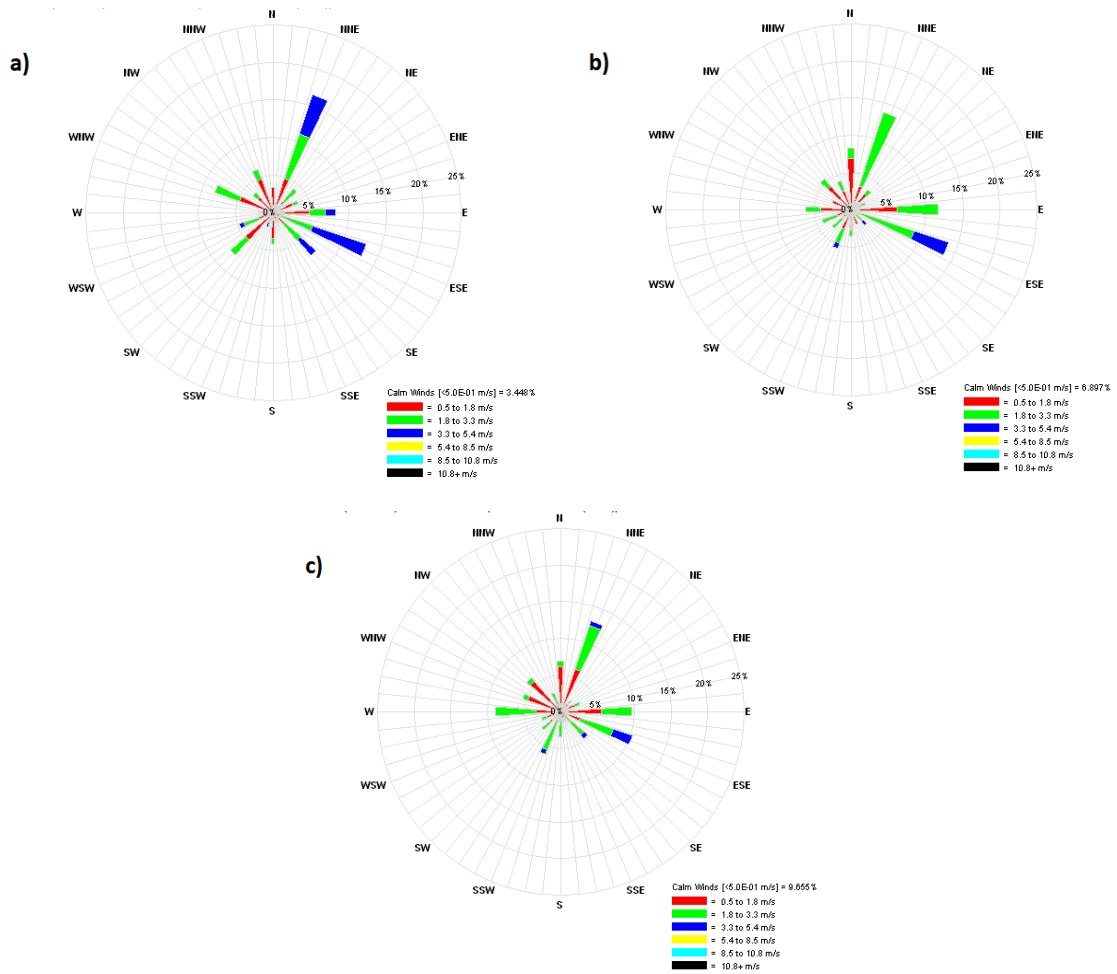
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2 Fig. 3 WRF domain grids resolution: D1: 27 km; D2: 9 km; D3: 3 km; D4: 1 km; D5: 333 m;  
 3 D6: 111 m (Cécé et al., 2016). D4, D5, D6 grids results provide the meteorological input to  
 4 FLEXPART and CALPUFF air quality simulations.

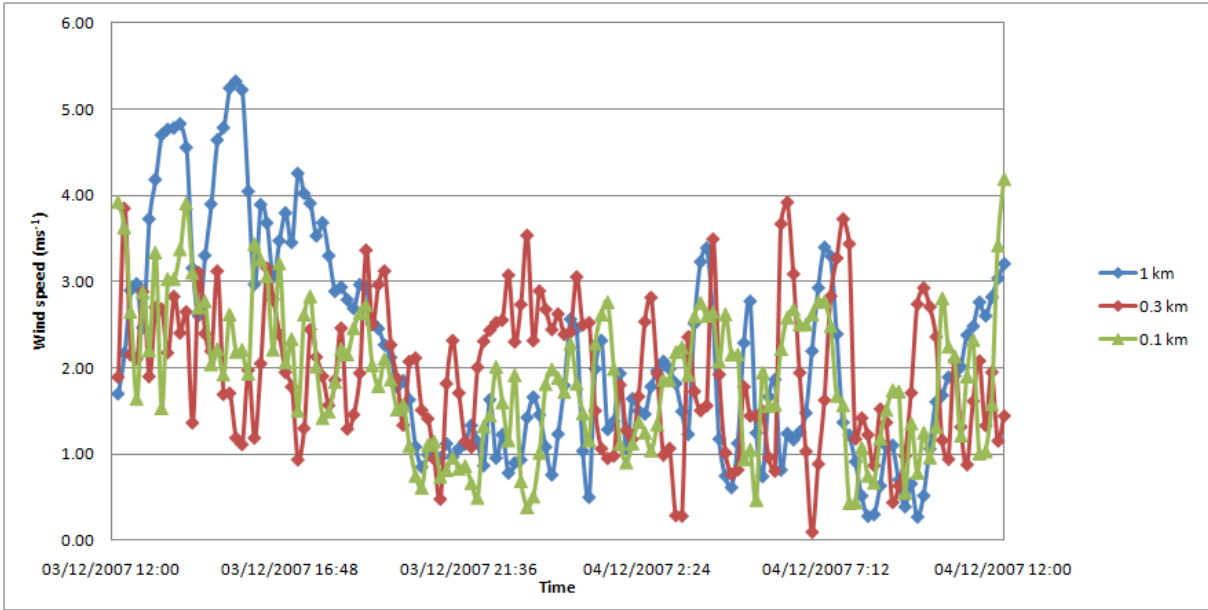
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2 Fig. 4 Comparison between wind roses at the emission source location from each WRF high  
 3 resolution grid output: a) D4: 1 km; b) D5: 333 m; c) D6: 111 m.

4



1

2 Fig. 5 Wind speed time series at the emission source location from each WRF high resolution  
3 grid output: D4: 1 km (blue diamond line), D5: 333 m (red diamond line) and D6: 111 m  
4 (green triangles line).

5



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2 Fig. 6 PWP plume observational pictures: a) longitudinal section; b) cross section.

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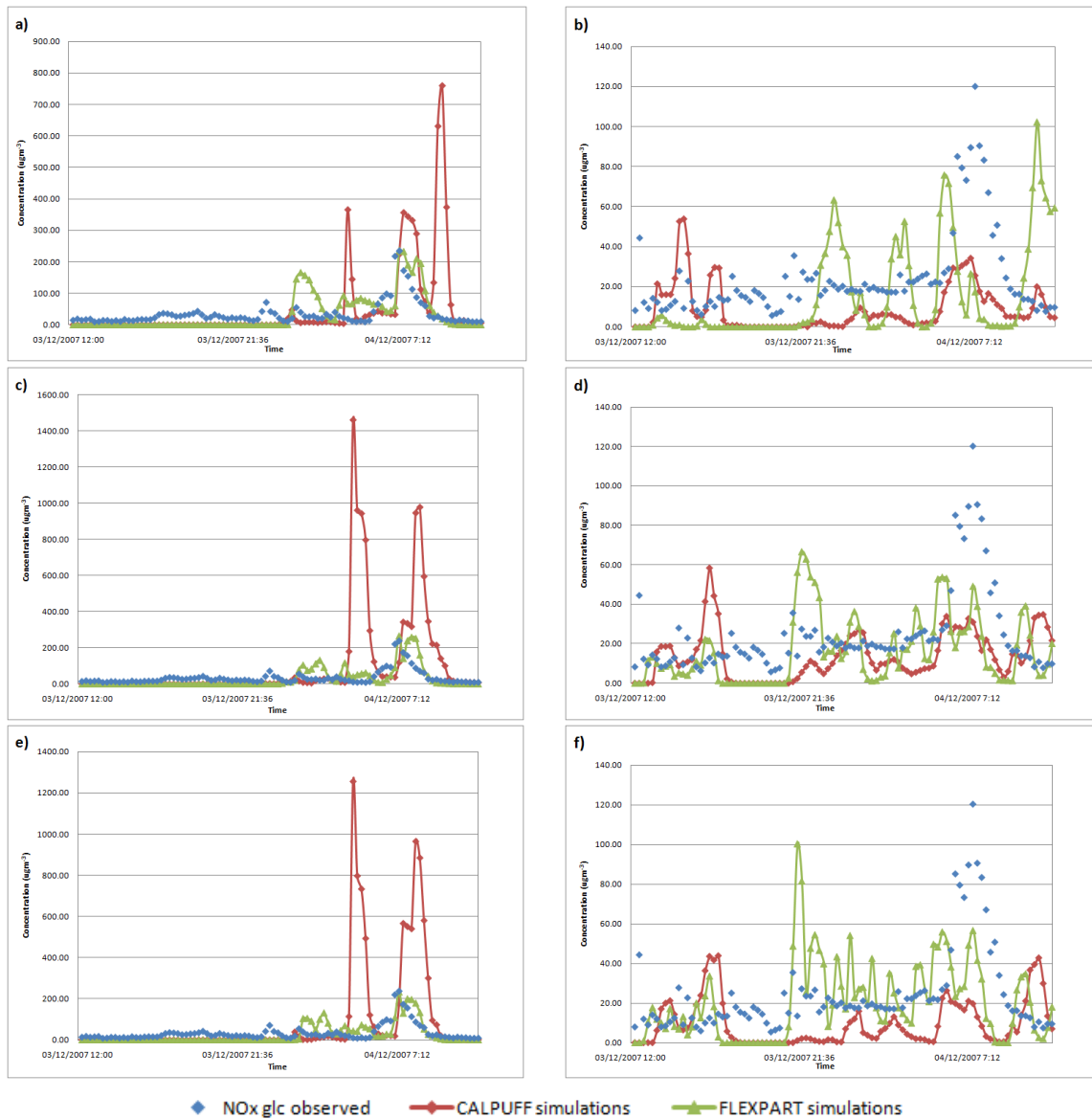
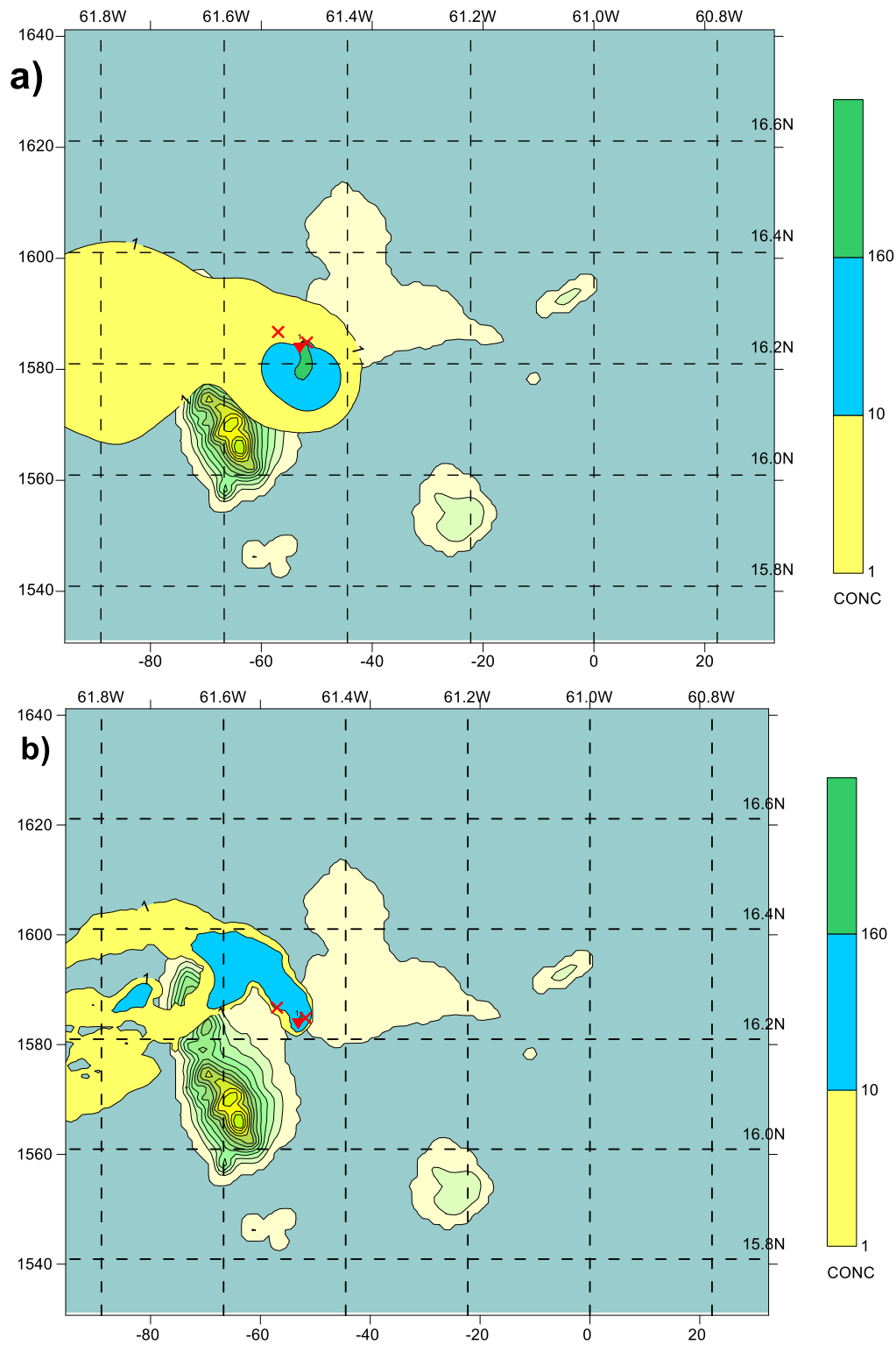
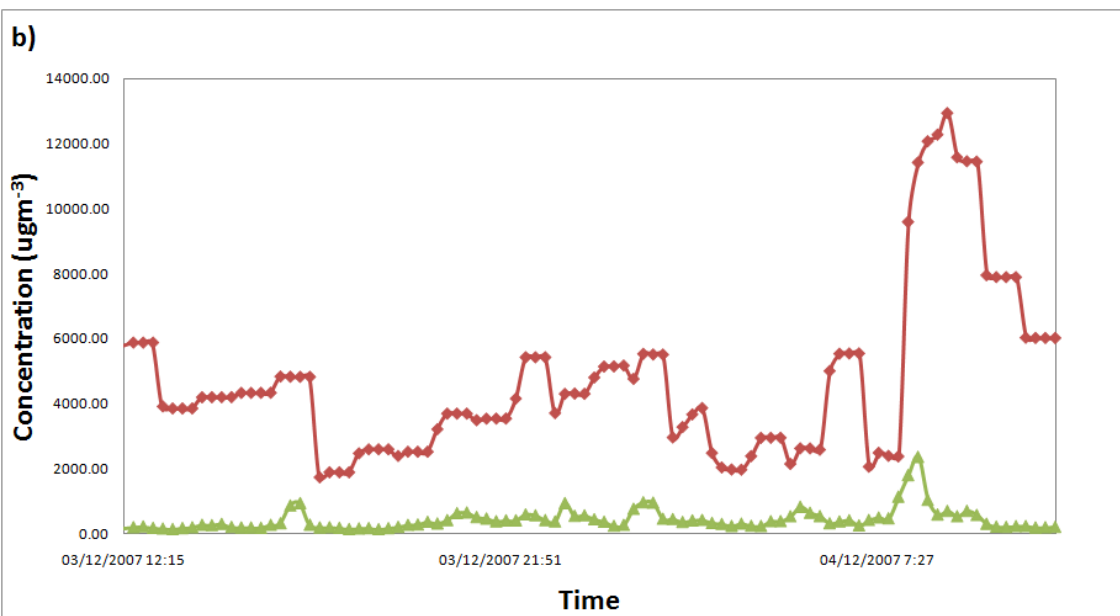
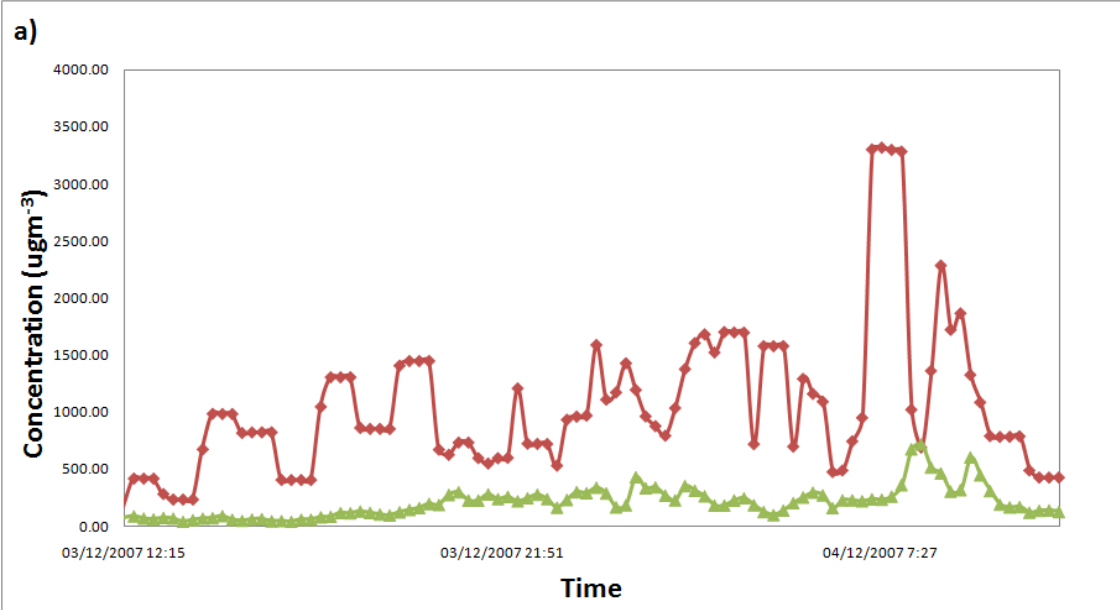


Fig. 7 Comparison between time series of 15-min  $\text{NO}_x$  glc ( $\mu\text{g m}^{-3}$ ) observed (blue diamonds) at AQS1 station (a, c, e) and AQS2 station (b, d, f), and estimated with FLEXPART (green triangles line) and with CALPUFF (red diamonds line) at the same stations, along the simulation period: 16 UTC 3 December 2007-16 UTC 4 December 2007; using different meteorological grid resolution inputs: a, b: D4 (1 km); c, d: D5 (333 m); e, f: D6 (111 m).



1 Fig. 8 Spatial distribution of  $\text{NO}_x$  ( $\mu\text{g m}^{-3}$ ) for 1 km of horizontal resolution domain (D4) at  
 2 05 UTC December 4, 2007 with source (red triangle) simulated with CALPUFF (a) and  
 3 FLEXPART (b). Also the emission source (red triangle), the two air quality stations locations  
 4 (red crosses), and topographic features are shown



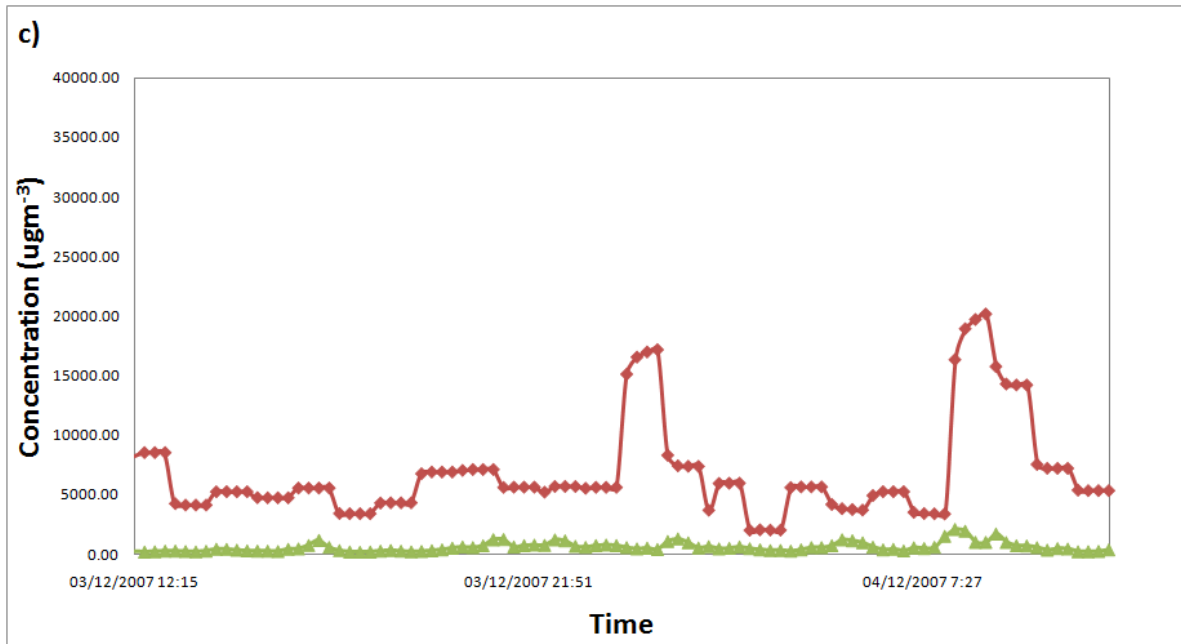
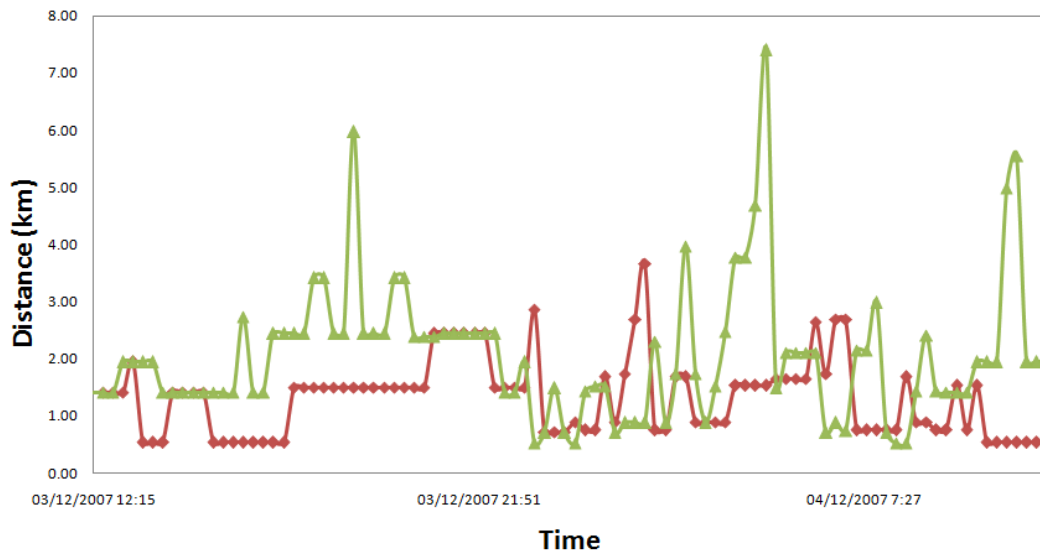
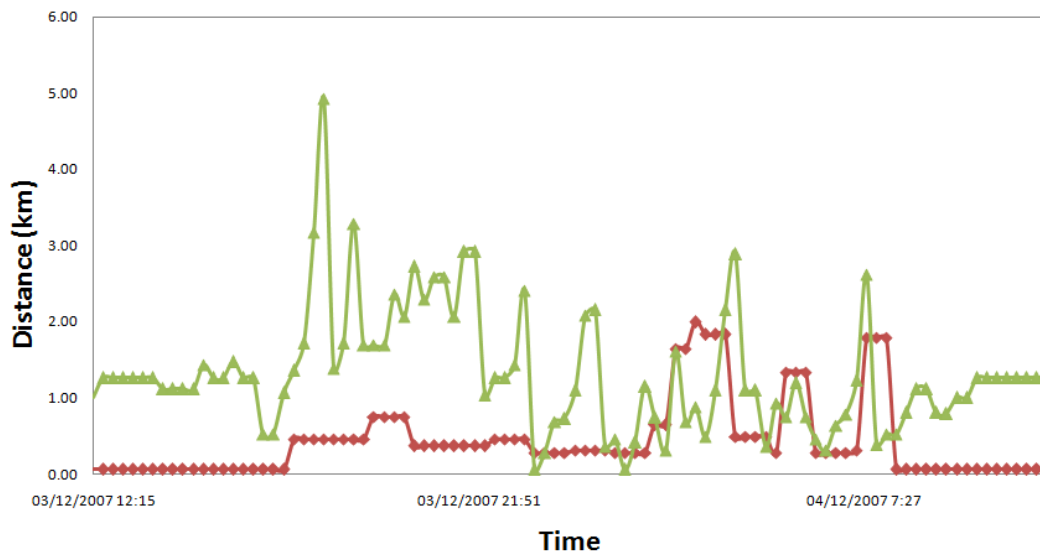


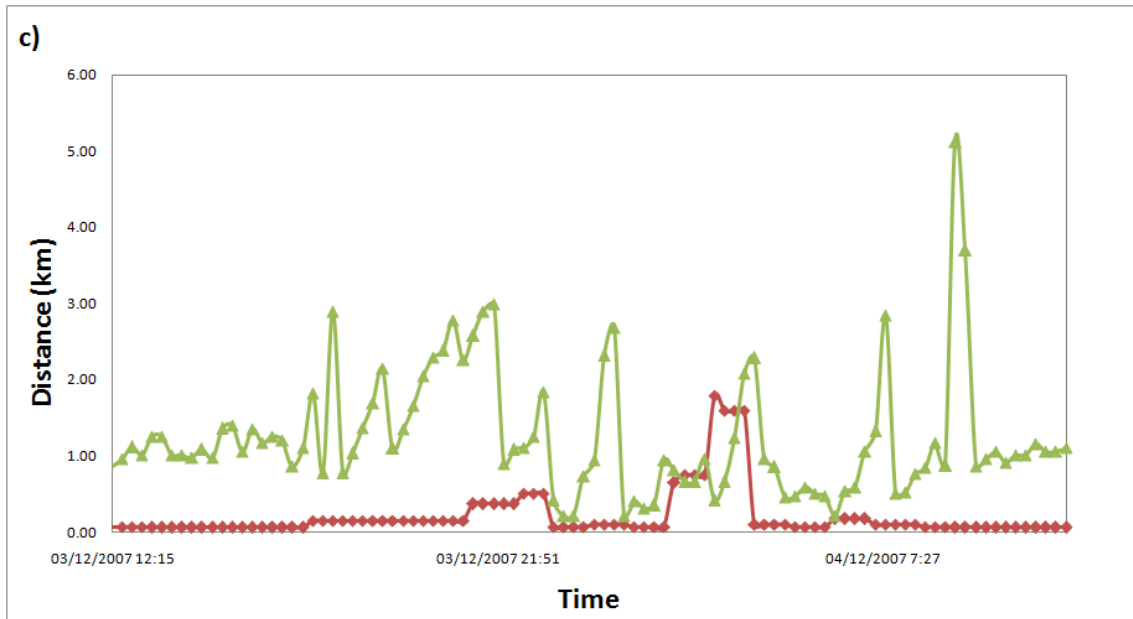
Fig. 9 Maximum NO<sub>x</sub> glc estimated with FLEXPART (green triangles) and with CALPUFF (red diamonds). Grid resolution: a) 1 km; b) 333 m; c) 111 m. Maximum simulated values for CALPUFF and FLEXPART respectively: a) 3 321 µg m<sup>-3</sup> and 716 µg m<sup>-3</sup>; b) 12 925 µg m<sup>-3</sup> and 2 369 µg m<sup>-3</sup>; c) 20 133 µg m<sup>-3</sup> and 2 092 µg m<sup>-3</sup>.

a)



b)





1 Fig. 10 Travel distance from the source to the maximum glc predicted with FLEXPART  
 2 (green triangles) and with CALPUFF (red diamond). Grid resolution: a) 1 km; b) 333 m; c)  
 3 111 m.  
 4

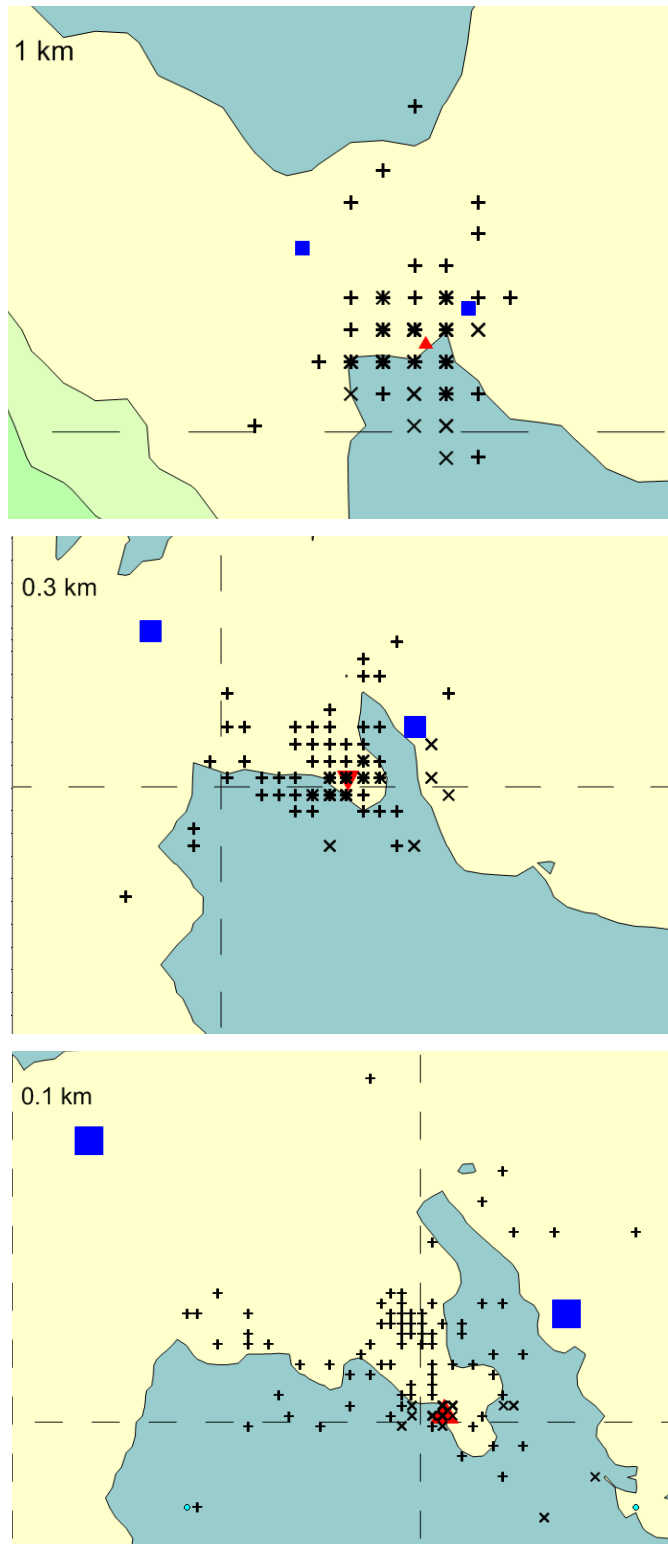


Fig. 11 Enlarged view of the maximum glc simulation locations using FLEXPART (+) and CALPUFF (x); also emission source (red triangle) and the two air quality stations (blue squares) locations are shown.

1 **Table I** Models' configuration.

Simulations	WRF grid resolution input	WRF Lowest vertical level height (agl-m)	PBL scheme
WRF/FLEXPART simulations			
flx1km	1 km grid resolution	10	Bulk Richardson approach from Voegelezang and Holtslag (1996)
flx0.3km	0.3 km grid resolution	No. of vertical levels	
flx0.1km	0.1 km grid resolution	38	
WRF/CALPUFF simulations			
Simulations	WRF grid resolution input	WRF Lowest vertical level height (agl-m)	PBL scheme
puff1km	1 km grid resolution	20	Ground: Bulk Richardson approach from Voegelezang and Holtslag (1996)
puff0.3km	0.3 km grid resolution	No. of vertical levels	
puff0.1km	0.1 km grid resolution	38	Overwater: Gryning and Batchvarova (2003) method

2 Note: FLEXPART and CALPUFF default options were applied, except above mentioned.

3



1 **Table II** BOOT statistics ( $\mu\text{g m}^{-3}$ ) for the simulation period, using FLEXPART and CALPUFF  
 2 results against observations at AQS1 and AQS2 stations.

Simulation	NMSE	FB	FBFN	FBFP
<b>AQS1 Pointe-à-Pitre</b>				
puff1km	8.97	-0.38	0.444	0.824
flx1km	1.79	-0.050	0.459	0.509
puff0.3km	20.17	-1.022	0.254	1.276
flx0.3km	1.89	0.093	0.540	0.447
puff0.1km	18.37	-0.905	0.331	1.236
flx0.1km	1.60	0.232	0.605	0.373
<b>AQS2 Baie-Mahault</b>				
puff1km	2.86	0.913	1.034	0.121
flx1km	2.94	0.429	0.852	0.424
puff0.3km	1.76	0.558	0.749	0.191
flx0.3km	1.49	0.387	0.630	0.244
puff0.1km	3.41	0.870	1.061	0.191
flx0.1km	1.23	0.165	0.483	0.319

1 **Table III** BOOT statistics ( $\mu\text{g m}^{-3}$ ) for the simulation period, using simplified CALPUFF  
 2 simulations and observations at AQS1 and AQS2 stations.

Simulation	NMSE	FB	FBFN	FBFP
AQS1 Pointe-à-Pitre				
puff1kmS	5.50	-0.240	0.448	0.688
puff0.3kmS	11.53	-0.821	0.284	1.105
puff0.1kmS	11.39	-0.581	0.397	0.978
AQS2 Baie-Mahault				
puff1kmS	5.30	1.317	1.335	0.018
puff0.3kmS	2.94	0.948	1.037	0.089
puff0.1kmS	7.83	1.382	1.434	0.052