We thank the referee for his or her constructive feedback, and time spent to review our manuscript in detail. Our responses to the referee's comments follow. The referee's comments are in italics.

Page 2 (lines 39-40): The emissions of 0.25 Pg C yr-1 proposed by Regnier et al. (2013) are based on Laruelle et al. (2010) and Cai (2011). Please include this information. See also Chen et al. (2013) and Laruelle et al. (2013), with updated values (0.10 Pg C yr-1).

> *Laruelle et al. (2013). Global multi-scale segmentation of continental and coastal waters from the watersheds to the continental margins. Hydrol. Earth Syst. Sci., 17, 2029–2051.*

We will add the appropriate reference to line 40 in the revised manuscript.

Page 2 (lines 42-43): Yes, and I agree. However, studies conducted in marine- dominated estuaries showed that they can present pCO2 values below 400 ppmv and a sink behavior. Please, check Koné et al. (2010); Maher and Eyre (2012), Cotovicz Jr. et al (2015).

> *Koné et al. (2009). Seasonal variability of carbon dioxide in the rivers and lagoons of Ivory Coast (West Africa). Estuar. Coast., 32, 246–260, 2009.*
>
> *Maher and Eyre (2012). Carbon budgets for three autotrophic Australian estuaries: Implications for global estimates of the coastal air-water CO2 flux. Global Biogeochem. Cy., 26,GB1032.*
>
> *Cotovicz Jr. et al. (2015). A strong CO2 sink enhanced by eutrophication in a tropical coastal embayment (Guanabara Bay, Rio de Janeiro, Brazil). Biogeosciences, 12, 5125-6146.*

Marine-dominated estuaries are briefly discussed in the Introduction in lines 91-99. Although they are underrepresented in data compilations, some behave as net CO_2 sinks. This will be explicitly stated in the revised manuscript and supported by the recommended references.

Page 3 (lines 76-78): Please check Borges and Abril (2011) to see details about the drivers of the emissions of CO2 to the atmosphere from estuaries.

Borges, A. V. and Abril, G.: Carbon Dioxide and Methane Dynamics in Estuaries, in: Treatise on Estuarine and Coastal Science, edited by: Eric, W. and Donald, M., Academic Press, Amsterdam, 119–161, 2011.

We thank the reviewer for the referral to Borges and Abril (2011). By analyzing a data set of nine European and two US estuaries, the authors concluded that 10 % of the total emission of CO_2 from inner estuaries could be attributed to the ventilation of riverine CO_2, whereas 90 % of the emission could be attributed to net heterotrophy. These two main drivers of estuarine CO_2 degassing are noted in lines 76-80 of this manuscript. Borges and Abril (2011) also detailed the relationship between freshwater residence time and the relative contribution of riverine CO_2 ventilation. This will be addressed in the revised manuscript.

Page 3 (lines 91-95): I think that the most overlooked estuarine typology regarding CO2 emissions are the marine-dominated estuaries.

We agree with the reviewer on this point.

Page 4 (lines 119-121): I would like to know about the wintertime period. Did you perform sampling campaigns at this period? Could you give some discussion about this?

With the exception of the 2016 cruise, water samples used in this work were obtained opportunistically on research cruises conducted during the spring and summer seasons. The Estuary and the Gulf are typically covered by ice during the winter months. Coast Guard icebreakers keep shipping lanes open during the winter, but, even though we have been offered berths onboard, the icebreakers will not stop for discrete water sampling. Hence, there is no data for the winter season. We have recently submitted, under a newly-created program, a multimillion-dollar proposal that would provide us with dedicated time on an icebreaker during the wintertime and, possibly, a vessel (CCGS Amundsen) equipped with an underway pCO_2 system.

Page 6 (lines 202-203): And also vertically, not?

The Upper Estuary is strongly laterally stratified, but because is it generally shallow and, due to the turbulence generated by tides, winds, and waves interacting with the bottom topography, it is only very slightly vertically stratified. In contrast, the Lower Estuary, because it is much deeper, is strongly vertically stratified. See the appended Fig. 1.
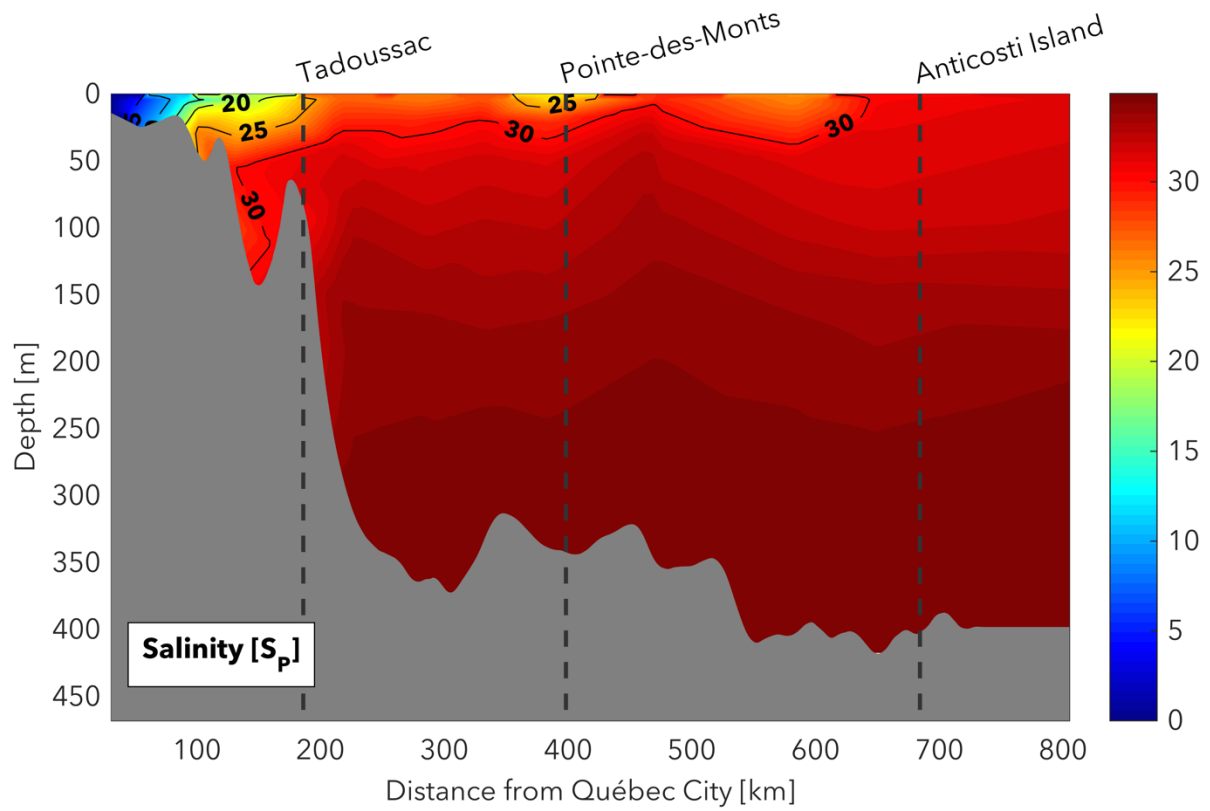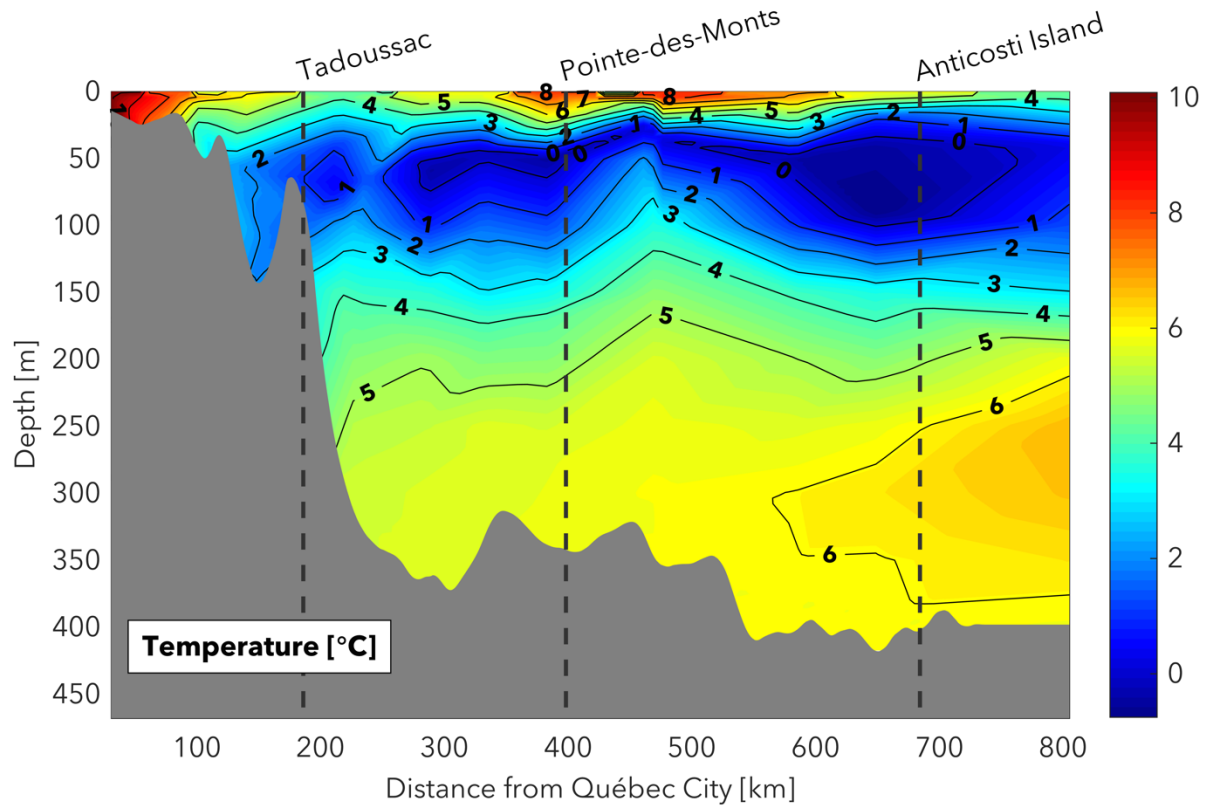
Fig. 1. Vertical profiles of temperature and salinity for the sampling transect during the May 2016 cruise.

Page 7 (lines 225-228): Please, provide more details about the sampling strategy. What is the vertical resolution (water column) of the sampling? How many samples did you take each year? How many samples did you take adding all campaigns?

As the present work focuses on the inorganic carbon chemistry of the surface mixed layer, the total number of surface-water samples (N) taken from the River, Upper Estuary, Lower Estuary, and Gulf are summarized in Table 2. Nevertheless, in all cases the whole water column was sampled, typically at 3 m, 20 m, 50 m, 70 m, 100 m and at 50m intervals to the bottom (or within 10 m of the bottom). These data will be presented in a subsequent manuscript in which the geochemical and isotopic characteristics of source-water masses and their relative contributions to the Estuary will be examined.

Page 8 (lines 240-244): Did you filter the water for the total alkalinity measurements? Previous works showed that for coastal waters the phytoplankton and bacterial cells can affect the measured alkalinity of unfiltered samples (Kim et al. 2006; Chanson and Millero, 2007). Do you have information about the particulate inorganic content (e.g., CaCO3) of the sampled waters? Please, see Kim et al. (2006) and Chanson and Millero (2007) about this problematic.

> *Kim et al. (2006). Contribution of phytoplankton and bacterial cells to the measured alkalinity of seawater. Limnol. Oceanogr. 51 (1), 331-338.*
>
> *Chanson and Millero (2007). Effect of filtration on the total alkalinity of open-ocean seawater. Limnol. Oceanogr.: Methods 5, 293–295.*

The sampling protocol we used is standard for TAlk and DIC measurements in open ocean waters (Dickson and Goyet, 1994). Hence, our samples were not filtered, but when SPM concentrations were high (in the turbidity maximum of the Upper Estuary), the SPM was allowed to settle for several days before the supernatant was sampled for analysis. Previous experiments with filtered and unfiltered samples revealed that results were identical within the analytical uncertainty (±2 µmol/kg) of our measurements.

The phytoplankton community composition in the study area is strongly dominated by diatoms (Devine et al., 2015). There is no significant population of pelagic carbonate-secreting organisms (coccolithophores, foraminifera, pteropods) in the St. Lawrence Estuary and Gulf. *Emiliania huxleyi* (coccolithophore) can be found at relatively low concentrations (maximum of ~10^7 coccoliths/L) in the upper mixed layer in the Gulf and

in the Strait of Belle Isle, while small specimens of *Limacina helicina* (pteropod) have been found in very low abundance (maximum of 18 individuals/m^2) in the Strait and in the Gulf (Levasseur et al., 1994; Cantin et al., 1996; Levasseur et al., 1997). Detrital carbonates are eroded from the Silurian-Ordovician deposits of Anticosti Island in the Gulf, but our sampling was carried out far from the source.

Dickson, A. G. and Goyet, C. (Eds.): Handbook of Methods for the Analysis of the Various 800 Parameters of the Carbon Dioxide System in Sea Water (Version 2), U.S. Department of Energy, 801 ORNL/CDIAC-74, 1994.

Devine, L., Plourde, S., Starr, M., St-Pierre, J.-F., St-Amand, L., Joly, P., and Galbraith, P. S.: Chemical and biological oceanographic conditions in the Estuary and Gulf of St. Lawrence during 2013, DFO Can. Sci. Advis. Sec. Res. Doc., 2015/013, 45 pp., 2015.

Levasseur, M., Keller, M. D., Bonneau, E., D'Amours, D., and Bellows, W. K.: Oceanographic basis of a DMS-related Atlantic cod (Gadus morhua) fishery problem: blackberry feed, Can. J. Fish. Aquat. Sci., 51, 881–889, 1994.

Cantin, G., Levasseur, M., Gosselin, M., and Michaud, S.: Role of zooplankton in the mesoscale distribution of surface dimethylsulfide concentrations in the Gulf of St. Lawrence, Canada, Mar. Ecol.-Prog. Ser., 141, 103–117, 1996.

Levasseur, M., Sharma, S., Cantin, G., Michaud, S., Gosselin, M., and Barrie, L.: Biogenic sulfur emissions from the Gulf of Saint Lawrence and assessment of its impact on the Canadian east coast, J. Geophys. Res.-Atmos., 102, 28025–28039, 1997.

Page 10 (lines-315-327): This paragraph is confused. As you said before "The samples were taken at 3 m...", then why no use this depth for the individual data points of surface-water pCO2 at each sampling location? It is not clear what data were averaged.

For each sampling station, data points in the surface mixed layer (SML), which exchanges heat and gases (e.g., CO$_2$) with the overlying atmosphere, were averaged. Given limited sampling in surface waters above the cold intermediate layer, the mixed-layer dataset at each station was most often limited to a single sample (from ~2 or ~3m depth), although, in a few cases, it included a sample taken at ~10m depth.

Page 10-11 (lines 331-348): This is an important section showing that your results at the low salinity region (0-5) can be overestimated or underestimated. This depends of the formulations of K1 and K2. Could you give more details about this? Did you perform

direct pCO2 measurements to compare results? What values of pH and TAlk did you use for the Figure 3? Please include this information in the figure caption.

As noted in lines 301-306, the pCO_2 values used in this work were calculated from the measured pH and TAlk, using the carbonic acid dissociation constants, K_1 and K_2, of Cai and Wang (1998). Whereas we have occasionally carried out DIC measurements on the water samples, direct pCO_2 measurements were not carried out as we do not have access to the proper equipment. We agree with the reviewer that direct pCO_2 measurements would have been desirable to confirm the accuracy of the pCO_2 calculations.

An underway pCO_2 system (General Oceanics model 8050) was operated by a colleague at the University of Manitoba, Prof. Tim Papakyriakou, and his students on the Canadian Coast Guard Ship (CCGS) and icebreaker Amundsen as it made its way through the Gulf of St. Lawrence to the Arctic for a scientific survey in early June 2016. Unfortunately, although the vessel left from its port-of-call in Québec City, the system was not turned on until well into the Gulf of St. Lawrence (near Anticosti Island) and through the Strait of Belle Isle. When in operation, the system was continuously sampling water from a high-volume inlet located at a depth of 5 m. Water was cycled through the underway system at a rate of 2.4 – 2.8 L min^{-1} and calibrations of the system's infrared gas analyzer (LI-COR model LI-7000) were monitored twice daily against three certified gas standards traceable to WMO standards. The underway system has an expected accuracy of 2 µatm (Pierrot et al., 2009). The underway pCO_2 measurements near Anticosti Island were in good agreement with the pCO_2 calculated in this study, for neighboring locations sampled in May 2016 aboard the RV Coriolis II (see the appended Fig. 2). Measured and calculated pCO_2 differed by, on average, ~4.2 %.

For the comparison of dissociation constants (results presented in Figure 3), values of pCO_2 were calculated at a constant temperature of 15 °C, at the measured pH and TAlk, using different published formulations of K_1 and K_2: Cai and Wang (1998), Lueker et al. (2000), Roy et al. (1993), Millero (2010), and Millero (1979) for pure water only. This is noted in the Figure 3 caption, lines 1163-1167.

Pierrot, D., C. Neill, K. Sullivan, R. Castle, R. Wanninkhof, H. Lüger, T. Johannessen, A. Olsen,R. A. Feely, and C. E. Cosca (2009), Recommendations for autonomous underway pCO_2 measuring systems and data-reduction routines, Deep. Res. Part II, 56(8-10), 512–522, doi:10.1016/j.dsr2.2008.12.005.
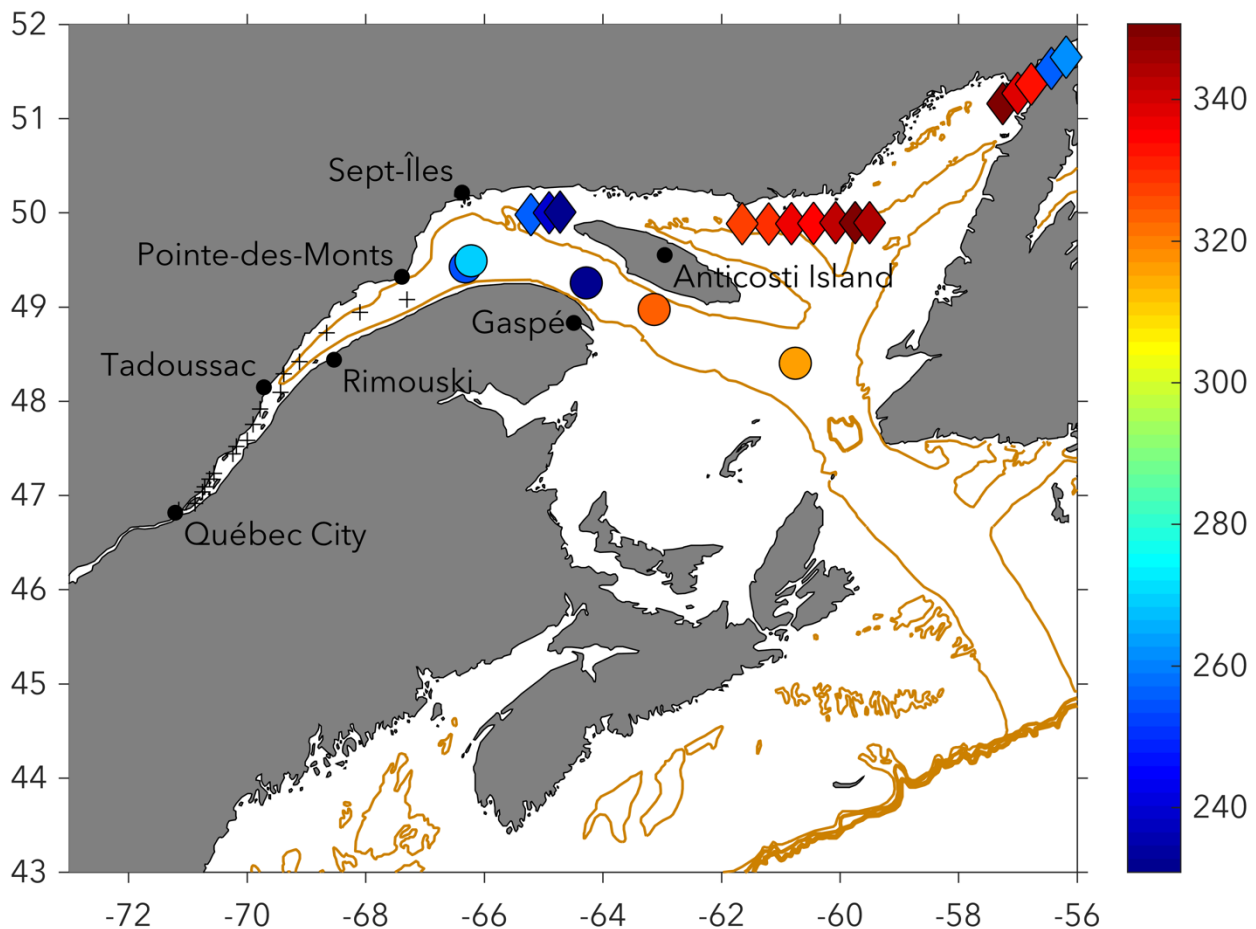
Fig. 2. Spatial distribution of surface-water pCO_2 (µatm) in the Gulf of St. Lawrence during Spring 2016. The circles show pCO_2 calculated from the data collected aboard the RV Coriolis II (May 2016), whereas the diamonds show pCO_2 measured from the underway system aboard the CCGS Amundsen (June 2016). The + symbols show all sampling locations during the May 2016 RV Coriolis II cruise.

Page 11 (lines 362-363) and Page 12 (lines 385): Could you describe the methodology for DIC analyses?

As noted in lines 301-306, most DIC values were calculated from the measured pH and TAlk, using the K_1 and K2 of Cai and Wang (1998). These calculated DIC values, along with the measured TAlk, were used to temperature-normalize the pCO_2, according to the method of Jiang et al. (2008).

Direct DIC measurements were carried out during the 2014 cruise using a Scitech Apollo DIC analyzer. After being thermostated at 25 °C, 1–1.5 mL of the sample was injected into the instrument's reactor where it was acidified with 10 % H_3PO_4 and the evolved CO_2 carried by a stream of pure nitrogen to a LICOR infrared analyzer. A

calibration curve was constructed using gravimetrically-prepared Na_2CO_3 solutions, and the accuracy of the measurements was verified using certified reference material solutions provided by Andrew Dickson (Scripps Institute of Oceanography). The reproducibility of the measurements was typically on the order of 0.2 %. Results of the direct DIC and pH measurements were used to assess the contribution of organic alkalinity to the total alkalinity in the Upper Estuary. The latter was found to be negligible (~–20 μmol/kg) relative to TAlk within the freshwater end-member (St. Lawrence River) and decreased seaward with increasing salinity along the Upper Estuary. Hence, measured and calculated DIC values were compatible. The DIC analytical methodology will be added to the revised manuscript.

Page 14 (lines 466-474): Do you think that hourly wind speed data averaged over the sampling month is a good approach to obtain a transfer velocity? Did you compare the hourly data average with other approach (minute/10minutes average wind speed) to investigate differences?

As the wind speed data used in this work were obtained from an external source (Environment Canada), we were limited to the data intervals of hourly, daily, or monthly. Average hourly wind speed data served as our best proxy for the steady (short-term) wind speed required by the equation of Wanninkhof (lines 460-461), which is applicable to estimate the gas transfer velocity from instantaneous wind speed measurements, using, for example, shipboard anemometers. The wind speeds provided by Environment Canada are observed from an anemometer.

It should be noted that shipboard measurements of instantaneous wind speed were available from the ship's log, but only for a limited number of stations from the 2010 and 2013 cruises. We elected to use the historical hourly weather data from Environment Canada for two reasons: (1) the data was more complete across time, and (2) the location of observing stations was consistent across space.

Pages 15 and 16 (section 2.6) : The approach of Carrillo et al. (2004) is a good way to compare the distributions and controls of biologically reactive dissolved gases. However, I also suggest compare the Apparent Oxygen Utilization (AOU) (Benson and Krause, 1984) with the Excess of DIC (E-DIC) (Abril et al 2003) that was applied in wide typologies of estuarine systems (e. g., Borges and Abril, 2011). Maybe you can compare the two approaches.

The DO percent saturation (%DO(sat)) values used in this study were calculated as: %DO(sat) = DO/DO* x 100, where DO is the measured DO and DO* is the equilibrium DO computed from the equation of Benson and Krause (1984). Similarly, AOU is

defined as: AOU = DO* – DO. Hence, we would expect the two approaches to yield similar results, and, in fact, they do, with the majority of data from the Upper Estuary showing the signature of respiration and the majority of data from the Lower Estuary and Gulf showing photosynthesis. See the appended Fig. 3.
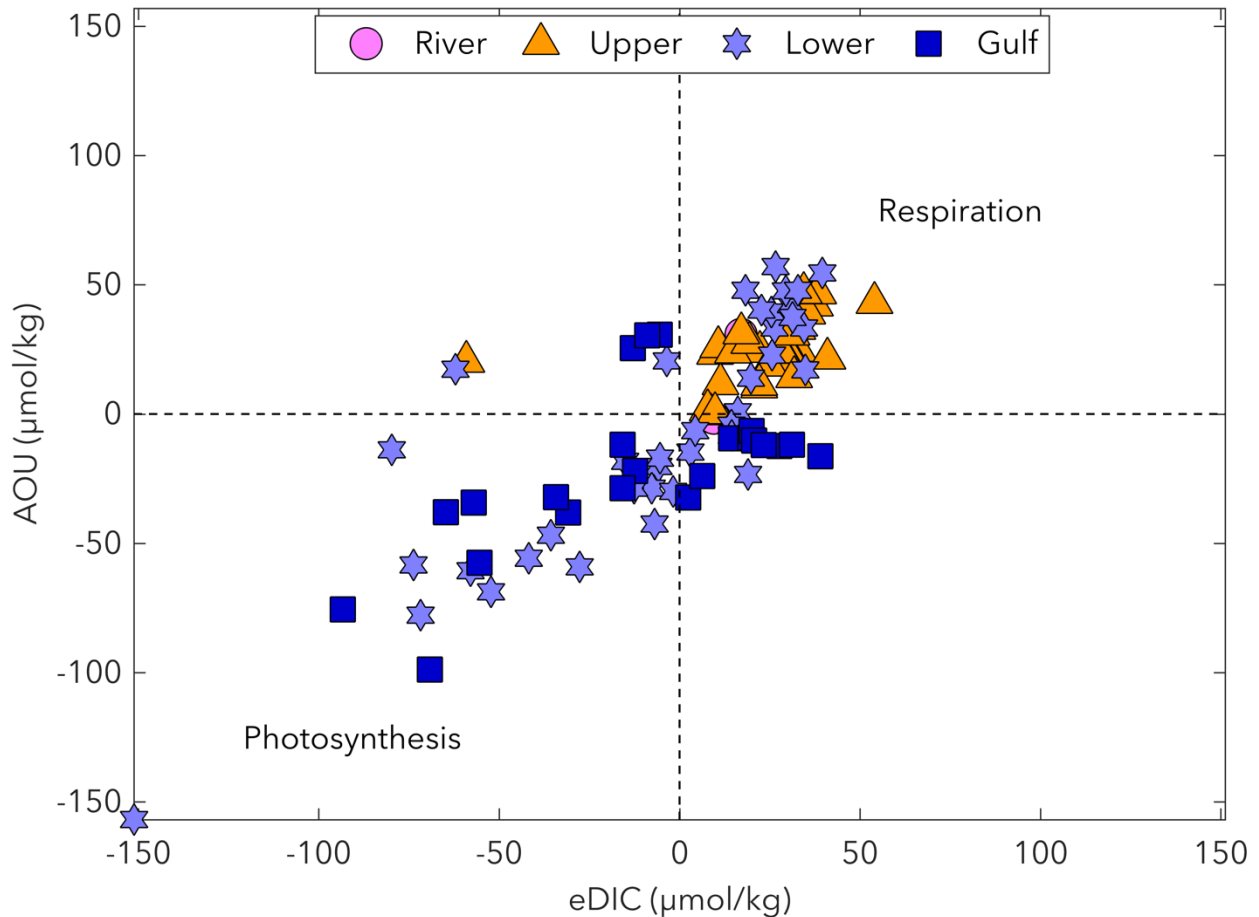
Fig. 3. Comparison of the apparent oxygen utilization (AOU) and excess DIC (eDIC) in the surface mixed layer waters of the St. Lawrence River, Estuary and Gulf. Positive values of AOU and eDIC indicate respiration, whereas negative values indicate photosynthesis.

Page 17 (lines 559-568): Do you have results of chlorophyll a concentrations? Do you have the results of column water stratification at this region? It's clear that the region of SLE near Pointe-des-Monts (between Tadoussac and Anticosti Island) present the pCO2 values lower than atmospheric pCO2 for all sampling campaigns. I think that some results of chlorophyll a and/or primary production could better support your discussion. I think you should include more discussion.

Whereas direct measurements of chlorophyll-a concentrations were not carried out during the research cruises, a fluorescence sensor was mounted on the CTD probe. Maximum fluorescence values measured in the euphotic zone at each sampling station are shown in the appended Fig. 4. Mean values of transmission (% light transmission) are also shown in Fig. 4. Low transmission values are due to light absorption by suspended particulate matter and colored dissolved organic matter, the latter being negligible in our study area. The maximum fluorescence values, along with high transmission values approaching 100%, were observed in the eastern Lower Estuary and western Gulf, where the system shifts from net heterotrophy to net autotrophy. Hence, these data further support our original interpretations.

The water column is strongly stratified in the Lower Estuary and the Gulf, which enhances biological production as discussed in lines 560-566. See the appended Fig. 1 for vertical profiles of temperature and salinity.
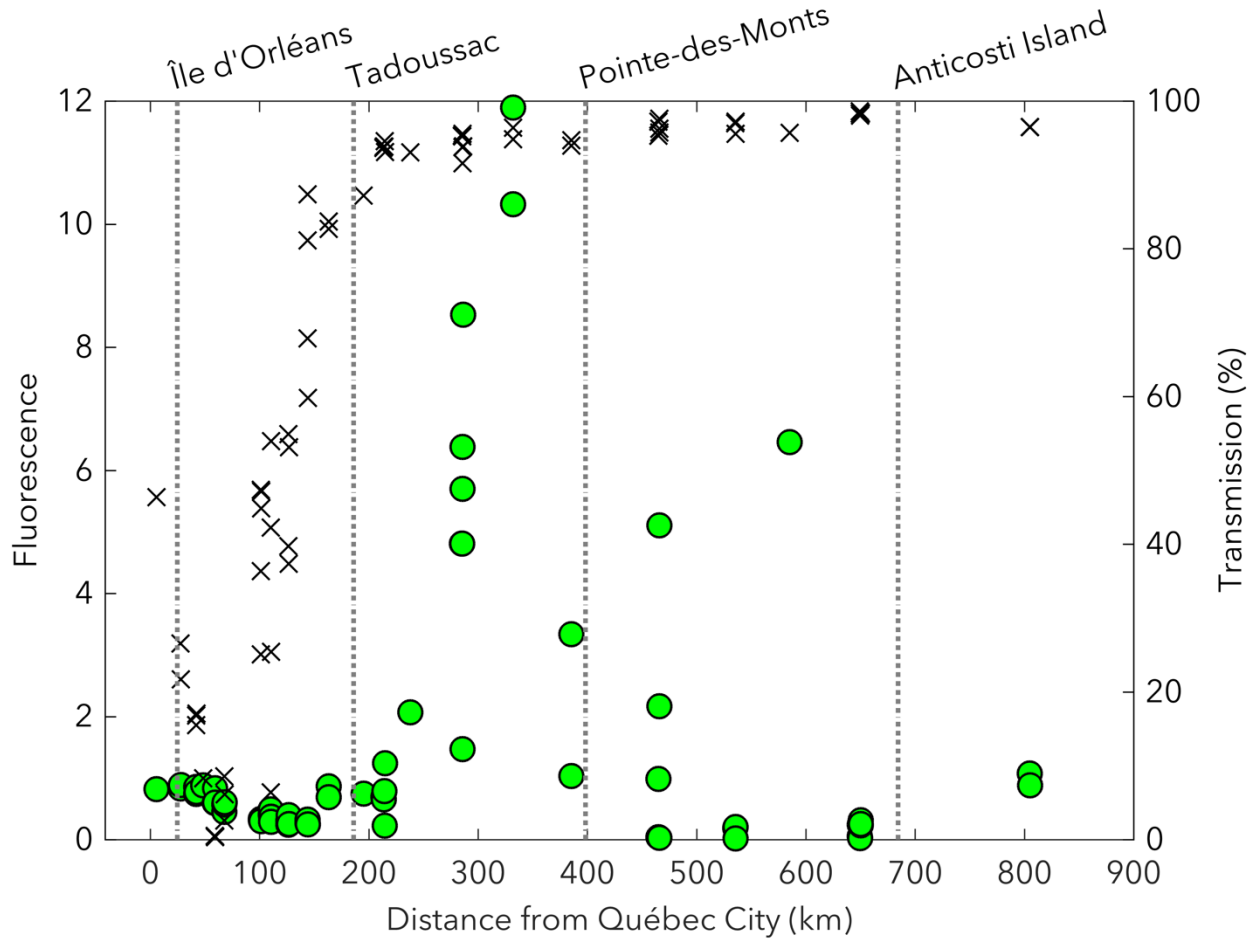
Fig. 4. Spatial distributions of maximum fluorescence values (circles) and mean transmission values (x symbols) in the euphotic zone in the St. Lawrence River, Estuary

and Gulf during the May 2016 cruise. Fluorescence is a primary production proxy, whereas transmission is an excellent proxy for turbidity.

Page 17 and 18 (lines 570-580): Could you give comparisons with other studies?

Hunt et al. (2014) indicate that temperature variability is one of the major controlling factors of seasonal changes in pCO_2 in the Kennebec Estuary, a large, river-dominated, macrotidal estuary. Had we been able to assess the temporal variability of pCO_2 in the St. Lawrence Estuary throughout the year (e.g., from summer to winter), we would expect that temperature exerts more control on the pCO_2 changes, due to larger seasonal temperature variability as well as the absence of phytoplankton blooms in winter.

Page 18 (lines 606-608): Previous studies identified CO2 sink behavior in estuaries. Please see Koné et al (2010); Maher and Eyre (2012); Cotovicz Jr et al (2015).

We thank the reviewer for bringing these studies to our attention as they demonstrate that some estuarine systems (e.g., those characterized by strong stratification and/or marine dominance) behave as net CO_2 sinks rather than CO_2 sources. We will refer to these studies in the revised manuscript.

Page 18 and 19 (Section 3.2): Please, provide comparisons of CO2 fluxes with other studies. Could you identify in the Figure 1 (or other) each segment considered in this study to the calculations of the area-averaged air-sea CO2 fluxes?

We agree with the reviewer's comment that the segmentation of the Estuary for the calculation of the area-averaged CO_2 flux should be shown on a map of the study area. See the appended Fig. 5, which will be used in the revised manuscript.
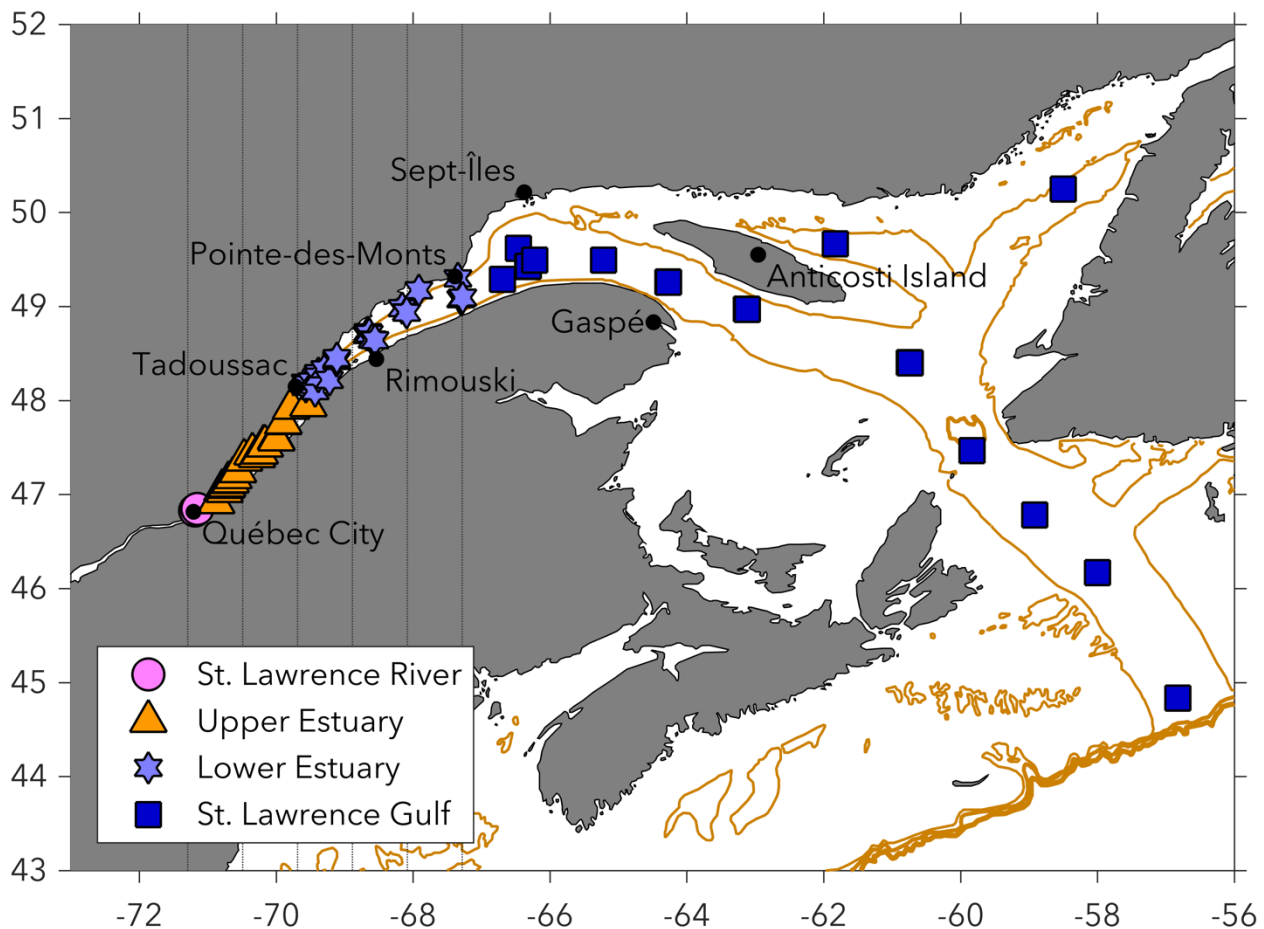
Fig. 5. Map showing the four principal subdivisions of the study area and the sampling locations (markers). The vertical dotted lines delineate the five segments of the Estuary used for the calculation of the area-averaged CO_2 flux.

Page 19 (lines 628-631): The biological activity can also drawdown the DIC concentrations in water column.

Photosynthesis is mentioned in line 628, but we agree with the reviewer's comment that the biological removal of DIC could be explicitly stated in line 629.

Page 20 (lines 645-651): In this section you can find other estuaries that acts as CO2 sink. Also, I think you could search in literature other papers that compared the biological x temperature effects over pCO2 concentrations to better contextualize you work (Bozec et al.,2011; Zhang et al., 2012; Hunt et al., 2014; Cotovicz Jr et al., 2015).

> *Bozec et al. (2011). Diurnal to inter-annual dynamics of pCO2 recorded by a CARIOCA sensor in a temperate coastal ecosystem (2003–2009). Mar. Chem., 126, 13–26.*

Zhang et al. (2012). Distribution and seasonal variation in the partial pressure of CO2 during autumn and winter in Jiaozhou Bay, a region of high urbanization. Mar. Pollut. Bull., 64, 56–65.

Hunt et al. (2014). CO2 Input Dynamics and Air–Sea Exchange in a Large New England Estuary. Estuar. Coast., 37, 1078–1091.

The relative importance of physical and biological controls on spatial variations of surface-water pCO_2 in the St. Lawrence Estuary will be more rigorously examined in a follow-up manuscript, by applying a quantitative water-mass analysis to remove the effect of physical mixing on the pCO_2 observations.

Figure 4: Could you move the legend of Figure 4 to outside the graph? Some points in the graph are over the lines of the axis. I suggest increase the ranges of the axis to put these points totally inside the graph. Also, it's evident that the GSL was sub-sampled compared to the USLE and LSLE regions. I think that it's important write about this in the methodological and discussion sections.

We agree with the reviewer's comment that the legend should be moved outside of the graph for improved readability, and the x-axis extended to encompass all data points.

The Gulf is sub-sampled in comparison to the Estuary because it is so large (~240,000 km^2 to the Estuary's ~12,820 km^2) and exhibits less variability.

Figure 9: Some points in the graph are over the lines of the axis. I suggest increase the ranges of the axis to put these points totally inside the graph.

We agree with the reviewer's comment that the x-axis should be extended to encompass all data points.

Table 3: I am not sure that the segments that you used to calculate the air-sea CO2 fluxes was the better division. Other graphs showed the results separated in river, upper, lower and gulf regions and maybe you can present the fluxes according.

The Estuary was divided into five equal segments to obtain a representative spatially-integrated whole-estuary CO_2 flux. Given that the Lower Estuary (Tadoussac to Pointe-des-Monts) occupies ~75 % of the total estuarine surface area, and encompasses a fairly wide range of pCO_2 values (standard deviation of 117 µatm), we decided to segment by

longitude rather than salinity to, more realistically, area-normalize the sectional fluxes (see Fig. 5).