BG 2016-380
Responses to Comments by Anonymous Reviewers 1 & 2

"Influence of infrastructure on water quality and greenhouse gas dynamics in urban streams'

We would like to thank the reviewers for their time in providing detailed, constructive comments regarding this manuscript. We have combined our responses to both reviewers' comments below, and believe that their contributions will lead to significant improvements. Both reviewers raised concerns about: 1) methodological details, 2) interpretation of results, and 3) terminology and clarity of ideas. Both reviewers outlined some overall comments with specific examples along with some technical line-by-line edits. As requested by the reviewers, we have combined our responses to each overarching comment with responses to specific examples where we deemed necessary below. We have not included direct responses to each line-by-line comment, but will incorporate these edits in the revised version of the manuscript.

I. METHODOLOGICAL INFORMATION

Both reviewers have expressed concern about the level of detail provided in the methods section, and certain specific methodologies used. We have compiled their general comments and replied to their examples where more than a simple textual response was deemed necessary.

R1 Comment 1: This paper appears to lack some methodological information, some of which is important and makes it difficult to assess what you did. Some of these examples of this are listed below.

And

R2 Comment 3) Some parts of the methods need clarification (e.g. supersaturation, DOM sample preservation). In addition some parts of the methods seem unnecessary given the results that are presented

In the submitted version of this paper, we described the different terminology for gas saturation in stream water (saturation ratio, or $xsCO_2$, $xsCH_4$, and xsN_2O) on page 7, lines 19-24.

In the revised version, we will add the following text to this section. "Super-saturation is defined as having a saturation ratio >1 or when $xsCO_2$, $xsCH_4$, or xsN_2O is >0."

We are not sure which aspects of DOM sample preservation the reviewer finds to be missing from the manuscript; however, we will clarify our preservation methods with citations below. We will additionally review our methods section again after completing

all other text edits to ensure that only the relevant methods are reported.

R2, P6, L16: 0.7micron-filtered samples stored for 2 weeks seems inappropriate for a DOM composition analysis. 0.2 micron filtering is usually preferred.

In the original manuscript, we describe DOM sample preservation and analysis on page 6, lines 16-21. Following filtration through pre-combusted 0.7μM glass fiber filters, samples were stored in amber glass vials at 4°C and analyzed within 2 weeks following collection. To the authors' knowledge, this is an appropriate and commonly utilized filtration procedure for DOM fluorescence metrics. Glass fiber filter pore sizes are not available below 0.7μM, and smaller filter materials (such as 0.2 μM nylon) have the potential to leach out fluorescently active compounds and/or measureable amounts of dissolved organic carbon during filtration.

Numerous references are available outlining this filtration and storage procedure (Sing et al. 2014, Sing et al. 2015, Huguet et al 2009, Dubchick et al. 2010, Gabor et al. 2014). While none of these papers specifically discuss the length of time that samples can be stored, the 'two week rule' is a commonly used convention rather than a biologically based limit to storage (Personal communication, Rachel Gabor, Shuiwang Duan). However, we can acknowledge in the revised text that some highly labile compounds can break down within hours of collection, while recalcitrant DOM can take months to break down contributing to some uncertainty.

R1: P4 L22-24 and Table 1: You may want to explain why you decided to treat these watersheds as four categories of two replicates each, rather than eight watersheds varying continuously along a few axes (impervious surface cover, development age, etc.). I think the reason of different discrete stormwater infrastructure design types going with developments built at different times makes sense; you just might want to state it a little more explicitly.

The reviewer's understanding of our reasoning for treating watersheds as replicates of different categories is correct. We do attempt to explain the reasoning for development of infrastructure types (page 4, lines 22-24), and we will clarify this section as follows:

" ... We selected eight headwater streams, each of which drained one of four distinct groupings of infrastructure types. Watersheds drained by these streams fell into four categories, which were based on development age, stormwater infrastructure design, and sanitary infrastructure. These headwater stream sites were treated as four discrete categories rather than eight sites across a gradient based on similarities in the form and age of stormwater and sanitary infrastructure in the watershed of each stream site. A comprehensive description of attributes in each infrastructure type can be found in Table 1; however, for simplicity we have abbreviated the types based on the dominant infrastructure feature as follows: 1)

stream burial, 2) in- line stormwater management (SWM) wetlands, 3) riparian/floodplain preservation, and 4) septic systems.

We will additionally review the remainder of the text to ensure that the infrastructure groupings are not described as a gradient.

R1: P4 L26-28: Over what time period (i.e. year(s), season(s)/month(s)?, times of day?) Actually, you should probably give much of this this information earlier than this section, and I don't think you did.

We will add the following information to the methods section of the manuscript (P4 L26-28)*: "Headwater stream sites were sampled every two weeks for both water chemistry and dissolved gas concentrations. Chemistry sampling took place for two years, between January 2013 and December 2014. Dissolved gas sampling took place every other week between July 2013 and July 2014. Sites were visited between the hours of 9am and 2pm."*

R1: P5 L5: How did you define a study reach? Approximately how long were study reaches? This information should come up in the previous section.

R2, P5, L5: Unclear what is meant by "study reach". It has not been defined.

Both reviewers pointed out the need to define the 'study reaches' established at each headwater stream sampling site. We will clarify our study design in the methods section as follows: "At each headwater stream site, we took five gas samples along a 20-m reach characterized by uniform bed forms either downstream or upstream of a stream gaging station. Gas samples were collected with 120-mL syringes at 0, 5, 10, 15, and 20m from the fixed starting point.

R1: P5 L17 & P8 L21: "Estimated using Google Earth software" sounds a bit sketchy. If you must mention Google Earth, include a citation for the program. Ditto at 8(21)), and also, what's the precision on the Google DEM, and why didn't you use the lidar one mentioned in 5(19-20); is it not more precise?

This analysis was originally completed using estimations of elevation and distance along the stream within Google Earth Software. Due to uncertainties in the precision of the Google DEM, as pointed out by the reviewer, we will re-calculate these values using a 2-meter resolution LiDAR – based digital elevation model procured by the Baltimore Ecosystem Study. This will remove the reference to Google Earth here.

R2: P5 L17-20: There are multiple ways to make these calculations; what actual

commands or tools did you use to do this?

We will clarify in the methods section that we used Hydrology tools in the Spatial Analyst toolbox of ArcGIS in order to delineate watersheds above each sampling point for our headwater sites, as well as longitudinal sampling locations. We mapped each sampled location using latitude and longitude and used these as pour points in the hydrology tools workflow. Because sampling points were always co-located with road crossings in this urban watershed, we were able to acquire the latitude and longitude of sampling sites using Google Earth software (Google Inc. 2009). Headwater stream sites were mapped based on the top of the 20m reach. Watersheds were delineated using a 2-meter resolution DEM created from LiDAR collected by Baltimore County in 2002. We first corrected the DEM for spurious depressions using the "Fill" tool in the hydrology toolbox. Next, we calculated flow direction for each pixel of this filled DEM raster. We then used the Flow Accumulation tool to evaluate the number of pixels contributing to each downstream pixel. After ensuring that each pout point was co-located on the map streams (i.e. areas with flow accumulation >500 pixels), we used the 'Watershed' tool to delineate the pixels draining into each sampled location.

R1: P8 L14, 17, & 24) & P9 L1-4: What is K_{20}? You did not previously explain what GT (from K_{GT}) means in general terms, so if that explanation was supposed to translate; it does not do so effectively. Ditto with K_{SF6} and plain K; are those at ambient temperature?

We will clarify our description of each equation accordingly. K_{20} is K normalized to 20C for a given gas. K_{GT} is K for a specific gas at ambient temperature. Equation (7) describes the relationship between K_{20} and K_T.

R1: P8 L20 You say you, "measure[ed] the change in elevation over a reach with a handheld GPS unit." Isn't elevation from GPS units usually rather unreliable? Describe the precision of your GPS unit.

GPS units were used to determine the location of two points along the stream network. The GPS points were mapped and the distance between points was determined using GIS tools. A 2-meter resolution DEM (based on 2002 LiDAR provided by Baltimore County Government) was used to estimate the change in elevation between the two points.

R1: P3 L28: Go ahead and be more specific than "water chemistry" if you can do so concisely.

We will change this to "were sampled every two weeks for dissolved carbon and nitrogen concentrations as well as and dissolved gases."

R2, P5, L1: Please specify what blanks are here.
We will add detail here that we collected three gas blanks by pulling 25mL of helium from the same tedlar bag used for headspace equilibration

R2, P5, L26: Not sure this equation and the associated text are necessary according to the results shown later.
Reviewer 2 is correct that we do not discuss the results from this mass balance calculation later on and it could justifiably be removed. We will remove this, as well as panels (e) and (f) from Figure 5.

R2, P5, L29: What about minor tributaries? Define better what you mean by major tributary.
We will clarify in the text that major tributaries were those contributing more than 5% of the discharge to the main channel at a given point along the stream network.

R2, P6, L10-12: Specify how TDN and DOC were analyzed.

We describe how DOC and TDN concentrations were analyzed on page 6 lines 9-14. We will clarify that 'TDN' was measured using the 'TDN' method, which consists of high temperature combustion in the presence of a platinum catalyst, and clarify that the Shimadzu instrument was a "TOC Analyzer." If the reviewer would be willing to provide more detail about which information is missing we would be happy to clarify the methods further.

R2, P6, L29: Why use a new name for this index if BIX is the name normally used?

We will replace 'index of autochthonous inputs' with 'BIX' throughout the manuscript.

R2 Comment 2) The role of external (non-in stream) and non-biological sources of GHG is not well considered in the manuscript. This may also make some calculations such as the index of aerobic and anaerobic respiration inaccurate.

R2, P7, L25 to P8, L11: This index seems controversial and needs clarifications. Not sure it can be really applied because apparently, it does not take into account external (non-in-stream) GHG sources and non-biological GHG sources.

Reviewer 2 is correct that AOU does not account for non-biological sources of GHGs. We will clarify this assumption about using the index on page 8, lines 10-11 where we define AOU. We will clarify that that AOU differentiates between aerobic CO_2 and CO_2 of anaerobic or abiotic origin (and not anaerobic vs. abiotic origin). By using this index without an additional metric for abiotic CO_2, we must assume that the proportion of abiotic CO_2 is small and invariant across sites and dates sampled. Richey et al. (1988)

justified this assumption with the following statement: "At ambient conditions (pH 6-7, alkalinity of 500-1000 ueq), with dissolved free CO_2 of 100-150uM or higher, the CO_2 produced through respiration remains primarily as dissolved CO_2. Thus ionic equilibrium reactions can be neglected." Richey et al (1998)'s justification is not valid in all cases for our study, as pH measurements varied widely from 4.81 to 8.9, and site-average CO_2 concentrations were lower than 100uM on 20 out of 152 sampling sites and dates, and alkalinity was not measured. CO_2 and pH were only both within this range on 36 out of 152 occasions. Among these observations, there remains a significant, positive linear relationship between xs CO_2 and xsN_2O ($p= 8.36 \times 10^{-15}$, $r^2 = 0.83$) across all sites. In the next version of this paper, we will repeat analyses that include AOU for this subset of samples.

We would like to clarify that this index does account for external (non-in-stream) CO_2 and O_2 sources, and this was our main reason for using the index. Regardless of whether CO_2 and O_2 are produced within the stream, in the soil, or along groundwater flowpaths, the ratio of these two gases within the stream will represent the relative abundance of CO_2 production to O_2 consumption along that flowpath. Richey et al. (1988) and Daniels et al. (2002) are two examples of freshwater-based studies that used this index to evaluate anaerobic CO_2 production in freshwaters.

R2, P11, L21: The term "anaerobic CO_2 concentration" seems erroneous. It does not make much sense. The same applies for anaerobic N_2O or CH_4 concentrations.

In the original draft of this manuscript, we define 'anaerobic CO_2 concentration' in the methods on page 8 lines 10-11 as follows: 'Anaerobic CO_2 concentrations were calculated as the difference between aerobically produced CO_2 (assumed equivalent to AOU) and measured CO_2 concentration.' Anaerobic CO_2, as we define it, is just CO_2 that was not produced by aerobic respiration, which could also be abiotic. In response to this reviewer's comment, we will change terminology of this CO_2 source to be non-aerobic CO_2. We would like to additionally clarify that AOU is not used for any other gases (CH_4 or N_2O) and we do not make mention to 'anaerobic N_2O' or 'anaerobic CH_4' because, unlike CO_2, these gases are not produced and consumed in direct proportion to O_2.

R2, P7, L23-25: Unclear. Please explain better how Cesc was estimated from SF6 additions.

*C_{esc} is introduced in our manuscript in equation 6 in our manuscript (K_{20}= Cesc * S * V). When this equation is rearranged to solve for Cesc, Cesc = S* V/K_{20}. K_{20} was calculated using measurements of SF6 off gassing conducted by Pennino et al. (2014). Briefly, because SF_6 is an intert gas, the loss of SF6 along a reach is proportional to the gas escape velocity (K_{20}). S (slope) at these injection sites was estimated using Google Earth imagery (though we will re-calculate this with 2m resolution DEM, based on Reviewer 1's comments) and V (velocity) was measured in the field. We will update the methods section with this added detail.*

R2: Table 1: I do not think so many decimals are necessary for most of these variables.

Table 2: "0.000" = "<0.001" or "<0.0001"?

We will make our reporting of p-values more consistent (<0.001) throughout the manuscript.

R2, Table 4: If some variables were log-transformed (e.g. logDOC: NO3), this should be indicated in the methods section.

We will add detail about log-transforming the DOC:NO_3^- ratio to the methods section.

II. STATISTICAL ANALYSES

Comment 2: In your statistical methods (section 2.4, "Statistical Analyses,") you execute a number of models (linear mixed effects, stepwise linear regression, etc., yielding all the results in Table 2 and 5) testing similar or related things. This may constitute a statistical multiple comparisons problem, i.e. increased chance of Type I error (https://xkcd.com/882). Consider either combining models (e.g. in a structural equations modeling framework or similar) or correcting for this risk of error. At the very least, try to combine your categorical and continuous variables for into a single model for each gas.

Reviewer 1 expressed concern about the statistical approach of using two modeling approaches to examine controls on each gas species citing that this approach seems redundant. The authors acknowledge that using two separate approaches for the purpose of predicting gas saturation values would increase the chance of Type I error; however, this was not the aim of our approach. The two models were used to examine first, whether or not there was consistent variation in gases across the categorical comparisons of watersheds, and secondly to examine whether or not gases could be predicted based on broader gradients in physical or chemical constituents that existed across all sampling dates and locations. In response to this reviewer's comments we have additionally incorporated a bonferroni correction to the p-value by dividing the 0.05 significance threshold by the number of models (6 models total, three for each gas), so that only tests with p< 0.0083 are considered significant. This does not change our results.

III. INTERPRETATION OF RESULTS

R1 Comment 3: Some interpretations of your results, most but not all minor, don't entirely make sense, or seem incomplete. For example:

R1: 12(16-17): Are you sure the "influence" is actually "indirect" on "biogeochemical processes in streams," or does the "indirect" part really only apply to GHGs? It seems like those things listed are directly related to biogeochemistry in general.

We will clarify here that, while watershed infrastructure was not a statistically significant

predictor of GHG saturation in streams, the gradients in DOC: NO_3^- that we found across all infrastructure types was strongly correlated with GHG saturation. We interpreted this to mean that infrastructure may directly influence DOC and NO3- loading to streams, and that this C:N stoichiometry is likely to be an important controller of GHG abundance downstream. This is not to say that GHGs produced within sewers, stormwater wetlands, etc., are not important, but rather that the strongest correlations exist with continuous dissolved parameters rather than categorical.

R1: P12 L23: Plain "nitrogen" or "*inorganic* nitrogen?"

We will change the wording here to 'inorganic nitrogen'

R1: P13 L9-10: "stoichiometric conditions more favorable for denitrification" would be a DOC: nitrate ratio closer to 1:1? If that ratio is different in incoming groundwater, wouldn't the N_2O:CO_2 ratio from that groundwater be correspondingly different as well?

We are not sure we follow the reviewer's question here, however we see the need here to clarify our interpretation of DOC:NO_3^- and CO_2:N_2O ratios.

DOC: NO_3^- stoichiometry is one way to examine whether biogeochemical conditions are favorable for one microbial process over another, as Taylor and Townsend (2010) describe in their in-depth metadata analysis of DOC: NO_3^- stoichiometry across a wide range of ecosystems. Helton et al. (2015) also provide a comprehensive review of the ways in which stoichiometry between inorganic N and organic C can be interpreted in various ecosystems. The implications of this stoichiometry at small spatial scales, such as the stream-groundwater interface of headwater streams, can be more complicated, however, and we agree with the reviewer that our interpretation could be explained more clearly.

As noted by Taylor and Townsend (2010), a DOC: NO_3^- ratio of 1:1 is ideal for denitrification, while DOC: NO_3^- much below 1:1 signifies conditions favorable for nitrification. While this ratio reflects the biogeochemical condition at the location/time the sample was collected, it is the result of processes occurring along the upstream flowpath. In predominantly groundwater-fed streams, for instance, heterotrophic denitrification may consume significant proportion of DOC along groundwater flowpaths of a septic plume, thus drawing down the DOC: NO_3^- of upwelling groundwater. Denitrification converts DOC to CO_2 and NO_3^- to N_2 and N_2O. Numerous studies have

shown septic plumes to have high concentrations of NO3- (e.g. Aravena et al. 1993). DOC concentrations are variable, but tend attenuate with depth in the aquifer and/or flow distance along the plume (Aravena and Robertson 1998; Pabich et al. 2001). For instance, Pabich et al. found high concentrations of DOC (>20 mg/L) in the upper part of a septic plume, with an exponential pattern of attenuation with depth. Consistently high NO_3^-- paired with attenuating DOC can result in a very low DOC: NO_3^- ratio by the time groundwater reaches stream. These conditions at the stream-scale are is more favorable for nitrification. Since nitrification is a chemoautotrophic process, consuming CO_2 while producing N_2O, we would expect to see a negative relationship, or no relationship between CO_2 and N_2O if nitrification were the dominant N_2O production pathway in a given watershed. Instead, we find positive correlations between CO_2 and N_2O in nearly all watershed sites (Figure 4a). We suggest therefore that denitrification may be producing N_2O in the groundwater in our septic-dominated sites, and drawing down DOC: NO_3^- along groundwater flowpaths. This interpretation remains hypothetical, however due to a number of biotic and abiotic processes occurring at the same time. Further work measuring solutes and gases along a groundwater flowpath is necessary to identify the mechanisms producing high concentrations of N_2O.

R1: P13 L24-25: You've made a big jump here, from relatively high emissions in certain places to "globally significant." Consider reminding your reader ("reminding" insofar as this should go in the introduction first; currently it's all just missing) what it would take for these locally high emissions to be globally significant- what's the relative global contribution of streams in general; how much of global streams is urban stream, etc. It might make more sense to think of the impacts of NO_2 emissions in the city in terms of local air pollution than global GHGs. You might also think about if your findings suggest anything new for general biogeochemistry, as opposed to just the GHG emission application.

Rather than focusing on global emissions, we will point out here, and in other parts of the manuscript, that diffuse emissions from urban streams constitute a previously unaccounted for source of N_2O and CH_4. It is currently unknown how significant this source is, although one study shows that, for N_2O, sanitary sewers could emit as much N_2O per capita as current estimates for secondary WWTP plants (Short et al. 2014). There is evidence that most of the N_2O-N found in these streams originates as wastewater, and our study adds insight into the magnitude and variability of biogenic gases in streams draining septic and sewer infrastructure.

We will also emphasize the point that greenhouse gas emissions from urban streams may

represent an important export pathway, for C and N from stream networks. Our results suggest that gaseous losses may need to be considered in urban watersheds from the perspective of mass transport and watershed C and N budgets.

Since we did not measure NO_2 emissions, we are unable to comment on whether or not streams are a source of that gas in our study sites.

R1: Missing: How did you analyze "longitudinal variability," or the effect of "distance from watershed outlet," on any of the response variables, i.e., the output of the method described in section 2.1.3? You make claims about the results of this survey in section 3.6 and display graphs derived from the data in Fig. 5, and then about the significance of these findings in 14(10-18). However, it's never apparent that you did more than eyeball that data to assess spatial trends. Moreover, my eyeballing does not match your eyeballing; I don't see Fig. 5 as reflecting the patterns you describe in the text.

R1: P14 L10-18: See comment after "missing," in Comment 1; it is unclear if you did a statistical analysis to support these claims.

The above two comments address our longitudinal study. We agree with Reviewer 1 that a more statistical approach to interpreting this data set is necessary. Our purpose for sampling gaseous and dissolved C and N along these watersheds was to determine whether or not the high N_2O and CH_4 saturation values found in headwaters was specific to headwaters or ubiquitous throughout the watershed. We will remedy the current lack of numerical interpretation as follows: For a given sampling date, we will compare the range and coefficient of variation in CO_2, N_2O and CH_4 saturation values in headwater sites to the range and coefficient of variation in main-stem sites. This will address the basic question that we set out to answer regarding spatial variability. We will additionally remove the water balance information from Figure 5, as this does not add any insight to our interpretation.

R1: P15 L17-18: "Variation in nonpoint sources and flowpaths" is not really an independent variable you tested; you don't know what in the watershed, but outside the stream, is driving anything, beyond a bit of inference about groundwater.

We will remove this sentence in the conclusion.

R1: Section 3.5 and Fig. 4b: Why do you think the slope directions of the lines in Fig. 4b so variable? Address this in discussion.

Overall, the relationships between CH_4 and CO_2 were much weaker and more variable than the relationships between CO_2 and N2O. We show this figure in part to demonstrate that N_2O and CH_4 do not behave similarly in relation to CO_2. In response to this reviewer's comment, we examined potential drivers of the ratio of $xsCH_4$: $xsCO_2$. Total dissolved N (TDN) was negatively correlated with $xsCH_4$:$xsCO_2$, and TDN concentrations explained 66% of the variance in this ratio, while DOC:TDN ratio only explained 53%. Differences in N availability across infrastructure categories may explain why the slope values and directions are so variable. One mechanism of this could be competition between NO_3^- and CO_2 as terminal electron acceptors during anaerobic respiration.

R1: Table 5: You never interpret your K_{20} results in the discussion.

We will add a brief comparison of our K_{20} values with the literature as follows. Our estimated K_{20} for O_2 spanned a wide range, from 1.0x 10-8 to 548. Raymond et al. (2012) performed a metadata analysis of all measured gas transfer velocities currently in the literature. In order to compare our values to theirs, we converted our K_{20} units from 1/day to m/day by multiplying K_{20} by water depth. Our calculated K_{20} (m/day) values span the full range of their metadata analysis (4.1 x 10^{-10} to 179 m/day), with 95% of our measurements falling on the low end (i.e. below 10 m/day). This lower end of the range reported by Raymond et al. (2012) is consistent with their result that gas transfer velocity scales with stream order, as our sites were located in first order streams. We provide detailed calculations of K_{20} in order to describe how GHG flux estimates were performed, however we decided not to discuss the K_{20} values in the discussion because we were not specifically interested in K_{20} as a variable on its own.

R1: P12 L26: Can you not distinguish (or at least venture an educated guess) between "C and N inputs and/or microbial metabolism," based on measurements/calculations of these gases individually, together with those of other gases?

We will add to the discussion section our speculation that the degree to which DOC: NO_3 in streamwater is driven by variations in C and N loading to the landscape vs. microbial processing along flowpaths depends on infrastructure. For instance, samples from streams draining septic systems had the lowest DOC:NO_3^- ratios, and we believe this is principally driven by the low starting C:N ratio of wastewater, and paucity of carbon sources that intersect the septic plume flowpath. On the other end of the spectrum, stream draining 'floodplain preservation' typologies also had newer development, and thus potentially reduced influx of low C:N sewage into streams. At the same time, these watersheds also had highly connected riparian banks with organic-rich soils at the stream-riparian zone interface may also be hot spots of NO_3- removal via denitrification.

R2: P2, L4: Land use can alter GHG emissions from streams not only through changes in drivers of stream metabolism. Changes in external GHG sources (e.g. groundwater inputs, soil leaching, point sources) and some geochemical reactions may also be important. In general, only part of GHG emissions from streams come from in-stream metabolism. This relevant aspect is not made sufficiently clear in this manuscript.

We agree with Reviewer 2 that land use can alter external GHG sources to the stream, along with changing in-stream metabolism. In the present form of the paper, we make mention of external GHG sources on several occasions, however we do not specifically attempt to differentiate between external vs. in-stream GHG production as we do not have data to back up this type of analysis. In terms of potential external sources, we mention the role of external GHGs via groundwater flowpaths in the introduction (page 2, line 26) referring to the buildup to GHGs in groundwater that is connected with wetlands, as well as in line 31 on the same page, referring to direct leakage of gas from sanitary sewer infrastructure. We also discuss the role of N_2O produced via denitrification or nitrification along subsurface flowpaths on page 13 (lines 8-12) in the discussion section. The use of the term 'watershed continuum' in this section and others refers to the suite of flowpaths (surface and subsurface) by which sources of GHGs from infrastructure and the landscape are connected to the stream. We acknowledge that this point can be made more clearly in the manuscript, and will edit the text accordingly.

IV. DESCRIPTION OF STUDY DESIGN

R1 Comment 4: You refer several times to a gradient or continuum of stormwater infrastructure, but you never elucidate the relationships between or ordering of the infrastructure types that makes them constitute a gradient or continuum. Explain, up front and early. For example:

R1: P2 L29: Is the "along the urban watershed continuum" significant? Does something change along this gradient about the effect of the wetlands, or do you just mean "in urban watersheds?"

We will change the language from 'along the urban watershed continuum' to 'in urban watersheds' here, as recommended by the reviewer.

R1: P1 L16: It is not immediately clear how these seemingly discrete categories constitute "a gradient of stormwater and sanitary infrastructure"- gradient along what axis, what variable?

We will clarify in the text here that these are indeed discrete categories of infrastructure, across which we found gradients in C:N stoichiometry, dissolved oxygen, temperature, etc.

R1: P3 L20-21: "Urban watershed continuum" again- is that just a way to refer to the stretch from the infrastructure in the headwater downstream a bit, or are the different kinds of infrastructure arranged along a continuum, or what?

The reviewer is correct that it is a way to refer to the flowpath from the infrastructure to the headwater downstream a bit. It is a term that explicitly incorporates infrastructure as part of the stream network in urban watersheds – the infrastructure/stream interface may play a significant biogeochemical role at a watershed scale in urban ecosystems. We have focused our study on understanding the role of urban infrastructure on greenhouse gas dynamics in urban waterways. A growing body of work has shown that nutrient and carbon loads to streams, as well as the biogeochemical processes within flowing waters is related to not only to land cover (% impervious surface, urban density, etc) but also urban infrastructure. Connectivity between runoff-generating water sources (groundwater, overland flow, shallow subsurface flow) and urban infrastructure (sewer lines, stormwater conveyance pipes, drinking water pipes, constructed wetlands, etc). is likely to influence not only the anthropogenic inputs of C and N to waterways but also the relative importance of biotic interactions on C and N removal along flowpaths. Kaushal and Belt (2012) describe a conceptual framework of how urban-impacted flowpaths may influence downstream export of nutrients as the 'Urban Watershed Continuum.'

R1: P5 L22: This is the closest thing to an explanation you've made so far, and it still doesn't really make sense.

In this section we are describing our sampling along the stream network. We will use the term 'stream network' here to clarify meaning. These specific changes are cited below.

V. VAGUE WORDING CHOICES

R1 Comment 5: You could improve this paper by reducing vague and occasionally careless diction. Sometimes this problem makes your meaning somewhat unclear. For example:

Both reviewers had concerns about some of the vague and unclear phrasing in sections of this paper. We respond here to their general comments as well as the specific examples from their line-by-line comments. Generally, we will clarify the key ideas underlying this paper in the introduction, provide more concrete details to back up statements about the literature, and link our interpretation of results more clearly to the figures and tables provided. The key ideas will be clarified in introduction include as follows:

R2: Title: I have the feeling that something is missing in the title. Maybe the word "of" before "urban"?

We will change the title to 'Influence of infrastructure on water quality and greenhouse

gas dynamics in urban streams'

R1: P2 L3-4: Consider fleshing out "globally significant" with some actual numbers? Also, if you have space, it might not hurt to explain very briefly how this impact of rivers and streams on GHGs was determined. It is unclear here whether the figures you cite include urban streams or not, and why. In other words, could knowing about urban stream GHGs make these fluxes more or less "globally significant?" Without this piece of information, it is unclear if all of the potentially contributing factors to urban stream GHG emissions that you describe in the rest of the paragraph are already accounted for in the currently accepted stream GHG numbers and you're just partitioning sources, or if you might revise the numbers on stream GHG fluxes as a result of this study.

We will flesh out the claim that streams and rivers emit globally significant quantities of greenhouse gases as follows: 'Flowing waters transport significant quantities of carbon and nitrogen from terrestrial ecosystems to the ocean. Along these flowpaths, rivers also emit significant quantities of biogenic gases. Inland waters, including rivers, lakes and reservoirs emit 1.2 Pg C yr^{-1} *of* CO_2*, equivalent to about half of the annual terrestrial carbon sink (Cole et al. 2007; Battin et al. 2009). Bastviken et al. 2011 recently estimated that inland waters emit 103 Tg* CH_4*-C* yr^{-1}*, the greenhouse warming equivalent to 0.65Pg* CO_2*-C* yr^{-1}*. Seitzinger et al. (2000) estimated that rivers, estuaries and continental shelves emit 1.6 Tg* N_2O *N* yr^{-1}*, which is equivalent to nearly half of all* N_2O *emissions from the ocean.'*

We will additionally clarify that some of these studies do take into account N_2O *emissions from urban areas indirectly, by using population to estimate N inputs to watersheds (Seitzinger et al. 2000). There remain significant uncertainties in 1) the amount of N entering waterways from urban areas, 2) the proportion of N that is converted to* N_2O *along groundwater and surfacewater flowpaths, especially in urban areas. These uncertainties may not be important at the global scale, but do impact watershed N budgets. For instance, Gardner et al. (2015) conducted a nitrogen input-output budget based on the difference between estimated anthropogenic N loading to the watershed and fluvial N export from streams. They found that outgassing of N (*N_2O *+* N_2*) from the stream accounted for all of the missing N.*

R2 P1, L17: Unclear what is meant by "watershed continuum". I think it would be more correct to speak about river network. This study focuses on the river and not on the whole watershed. This should be clear throughout the manuscript.

R1 P3 (20-21): "Urban watershed continuum" again- is that just a way to refer to the stretch from the infrastructure in the headwater downstream a bit, or are the different kinds of infrastructure arranged along a continuum, or what?

These two comments are related to the term 'urban watershed continuum.' We agree that, as presented in this paper, the urban watershed continuum is not clearly defined.

We will clarify in the introduction that this term is meant to describe expanded connectivity between infrastructure, landscape, and streams, which can influence biogeochemical functions (particularly at infrastructure/stream interfaces). We will additionally clarify throughout the paper which scale of connectivity we are referring to in order to more precisely present our findings.

R1: P1(27-29): Your concluding sentence is rather vague; for a start, "influenced" could mean almost anything. Could you be a bit more specific about what the "influence" was and what the "implications" are?

R2: P1, L29: This last sentence of the abstract does not seem appropriate. It refers to emissions, which are not the focus of the manuscript. I would rather include a more conclusive sentence here.

These two comments refer to the final sentence of the abstract. We agree with the reviewers that our study does not focus on emissions and will remove the last part of this sentence starting with 'with significant implications…'

R2: P3, L20-24: Yes, but how much do streams contribute to whole watershed GHG fluxes?
R1: To put your results in context a bit better, see Gallo et al. 2014 ("Physical and biological controls on trace gas fluxes in semi-arid urban ephemeral waterways" in Biogeochemistry 121(1) pp.189-207). They did related measurements in ephemeral streams in urbanized deserts, with similar results. For just nitrous oxide emissions from urban streams, there are several more relevant papers; try searching "nitrous oxide urban stream," in Web of Science if you can. (No, I am not Gallo et al.)

While it is beyond the scope of this manuscript to robustly quantify emissions from streams in this region we acknowledge that more context is necessary here to justify the scalability of our results. We will incorporate a 'back of the envelope' scaling exercise based on 1) range of flux estimates, 2) estimate of stream surface area and 3) soil GHG emissions from ongoing work at the Baltimore Ecosystem Study (Groffman et al. 2000; Groffman et al. 2009; Smith et al. in prep), and 4) a recent estimate of anthropogenic GHG emissions in Baltimore County (Brady and Fath 2008) in order to place stream GHG emissions in context with other notably larger sources. We will additionally note that, while GHG emissions from flowing waters are small compared to other anthropogenic GHG sources, they are clearly linked to water quality. It is therefore important to note that updating infrastructure may provide the dual benefit of improving water quality and reducing GHG emissions. We will certainly also incorporate citations to Gallo et al. (2014), as this study is highly relevant to the growing understanding of greenhouse gas production in urban aquatic environments.

R2, P15, L25-28: I suggest the authors try to include more results-based conclusions and implications at the end of the paper. It also seems confusing that the authors emphasize wastewater here, when the paper is about streams and GIs.

The paper examines both stormwater and sanitary infrastructure. We will clarify that we think our results present evidence that N loading and GHG emissions are related to sanitary infrastructure. We will introduce this idea a bit more clearly in the introduction and discussion.

The following two comments refer to the same two sentences in the abstract:

R1: 1 L22: "These variables" refers to the "drivers of GHG dynamics," "infrastructure categories," or both? If it's the former, I guess this line just verifies that "nitrogen stoichiometry" etc. *are* in fact "drivers of GHG dynamics" in this context (as expected); if "these variables" are the "infrastructure categories," then it's a much more novel finding.

R2: P1, L23: Not sure these r^2 values are helpful here. It is not clear which statistical test was used.

These two comments refer to the same two sentences in the abstract. On page 1, line 23, we are referring to the relationship between drivers of GHG dynamics (meaning the previously listed variables: C:N stoichiometry, dissolved O_2, dissolved nitrogen, and temperature) and N_2O CO_2, and CH_4 from linear mixed effects models. We will clarify our meaning in the text as follows:

'While categorical analysis of infrastructure type vs. GHG saturation did not show significant differences among the pairs of watersheds, watersheds draining different types of infrastructure did yield strong gradients in continuous variables such as C:N stoichiometry, dissolved oxygen, dissolved N concentrations, and water temperature. Taken together in linear mixed effects models, these continuous variables explained 78%, 78% and 50% of variability in N_2O, CO_2, and CH_4 respectively.'

R2: P1, L26: Again, unclear use of r^2 value.
This line contains an error, as we did find significant differences in the relationship between CO_2 and N_2O amongst infrastructure categories. The line will be changed to reflect the different r^2 values the relationship between N_2O and CO_2 for each infrastructure category.

R1: P2 L16-17: When you talk about GI here, are you proposing that all GI will have the same effects, at least in terms of direction of change in GHGs, or might effects differ depending on GI type?

We will change the wording here from 'GI' to 'constructed wetlands and riparian

preservation' because that is what we are referring to here. We mean to hypothesize here that both of these practices may potentially increase CH_4 while reducing N_2O production and emissions from streams.

R1: P2 L20: "Source of uncertainty" for what? Do you just mean "uncertain," or do you mean that this role could change our understanding of global fluxes from rivers, or what?

We will change this sentence as follows: "Despite considerable funds spent on restoring aging infrastructure and improving water quality in cities globally (Doyle et al. 2008), the role of urban infrastructure on in-stream GHG emission remains under-studied."

R1: P3 L10: Specify *anaerobic* nitrification; this is unclear until 12(29). With plain "nitrification," it at first seems like N_2O must be a typo for NO_2^-. You also need a source here for the description of nitrification; I don't think Taylor and Townsend 2010 suffices.

We are not sure we understand the reviewer's comment here, as we do not use the term 'anaerobic nitrification' in this paper. As described on page 3, lines 9-10, nitrification is a chemoautotrophic process, which oxidizes NH_4^+ to NO_3^-. CO_2 is consumed during this process, and N_2O is also produced as an intermediate in the NO_3^- oxidation process. We have cited Taylor and Townsend (2010) because they provide an excellent framework for determining whether an environment is more favorable to nitrification over denitrification based on the ratio of NO_3^- to DOC. We will add a more general reference about nitrification in aquatic systems to the text (Schlesinger 1997).

R1: P3 L18: "GHG emissions"- what about them? "Increased GHG emissions?"

We will change the wording here to "Increased GHG emissions"

R1: P12 L27 & P15 L13: Provide a citation for "'hot spots'" if you're going to put it in quotes, so we can verify which definition of "hot spot" you mean. Also, decide if you're going to say, "'hot spot'" or just "hotspot;" be consistent.

Upon reflection on Reviewer 1's suggestions about this analysis, we plan to remove the term(s) 'hot spot' in this section the paper and consistently separate it into two words where we do use the term.

R1: P15 L23: "Role" or "influence?" Sometimes your point could be stronger if you provided concrete numbers to back up your assertions. For example:

R1: P2 L13: What does "substantially" mean? Can you provide numbers as to the relative contributions of nonpoint and point sources?

We will change this sentence as follows, by moving some background information from the discussion into the introduction. "Several studies have documented that wastewater leakage from municipal sewers often accounts for more than 50% of dissolved N in urban streams (Kaushal et al. 2011; Pennino et al. 2016; Divers et al. 2013). While sewer lines are known to leak dissolved N, N_2O losses are not accounted for in greenhouse gas budgets of large WWTPs that these pipes feed into. Short et al. (2014) measured intake lines from three municipal WWTPs and estimated that N_2O emissions from gravity sewer lines alone on the same order of magnitude (1.7g N_2O person yr^{-1}) as current IPCC estimates for per-capita emissions from secondary WWTPs. Their study demonstrates the importance of constraining biogenic gas emissions from streams which flow alongside aging sewer lines."

R1: P3 L22-24: How is human population relevant? Also, please contextualize "fastest form of land use change;" that statement alone isn't really enough to ascertain significance. Is the magnitude of the change (i.e. first derivative of land use rather than second derivative) large? Is urban land use large, relative to other uses? Or do you think urban watersheds contribute disproportionately much to GHGs for their size, and so are significant globally even if small?

We will remove this sentence and clarify as follows:

'Our study investigates patterns in GHG abundance and emissions from urban streams. This source of GHG emissions remains poorly constrained due to 1) heterogeneity of aquatic ecosystems within urban watersheds, and 2) uncertainties in emission factors (i.e. the percent of N added from a particular source that becomes N_2O) due to a range of N sources in urban streams (wastewater, atmospheric deposition, fertilizer). All of these sources, but especially wastewater, tend to increase with population rather than land cover explicitly. Wastewater N loading to rivers is projected to more-than double between 2000 and 2050 (van Drecht et al. 2009). While wastewater currently only comprises about 3% of anthropogenic N_2O emissions globally (IPCC 2006), Strokal and Kroeze (2014) demonstrate that increasing population and thus N loading will almost certainly lead to higher N_2O emissions, regardless of increased water treatment. It is therefore crucial to evaluate the ways in which highly managed urban watersheds process excess N and produce GHGs.

R1: P12 L6: Which were the "three high-flow sampling dates?" Sometimes you waste

valuable space by not going ahead and saying what you actually mean.

W*e will list the three dates when high-flow conditions were sampled here.*

R1: P2 L9: Again, on "implications," try to be less vague if you can do so concisely. "Increase or decrease" or "change the magnitude of?" "Alter seasonality of?" Etc.

R1: P15 L1: By "typologies" you mean "types?"

In the methods section (page 4, lines 20-24), we describe the combinations of sanitary and stormwater infrastructure in each pair of similar watersheds as 'typologies.' This could be changed to 'categories' if this helps to clarify our categorization of these watersheds based on infrastructure types.

VI. TRANSITIONS, DEFINING TERMS, ETC

R1, Comment 6: Remember to maintain coherence and clarity of the paper through clear transitions, linking similar ideas, defining terms the first time you mention them, etc. For example:

R2 Comment 1) Some strange terms are used throughout the text that could be avoided (e.g. "watershed continuum", anaerobic concentration)

We have taken these reviewers comments into consideration and will make changes to the wording, definition of terms and transitions of ideas throughout the manuscript. We have included examples of our response to these concerns below.

R2: Abstract: You don't describe your "longitudinal" results here (the ones along stream length).

Following the changes to our statistical analysis and interpretation of these results that we have described above, we will add a sentence to the abstract describing differences in the variance GHG saturation in headwater streams, compared with main channel sites.

R1 P2 L21-23: How do these numbers/methods for calculating global fluxes that you cite here compare to the ones in 2(2-3)?

R1 P3 L29 - P4 L2: The final sentence in this paragraph seems out of place. Maybe shift it to the start of the next paragraph and end with, ", which facilitated site selection," or something? If you don't move the sentence, at least go ahead and explain why this information store matters. I mean, I can guess, but I shouldn't have to do so, or to wait

until you bring it up again later. Maybe just collapse the first two paragraphs into one?

We will remove the last sentence of this paragraph.

R1 P4 L5-6: Clarify timing. Everything was put in place in the 1950s-1970s, and the aging and cracking is now (or rather, when this study was conducted)? Also, "between" or "from?"

R1 P4 L13: Remind us *which* eight streams- "...the eight streams *studied* drained...?"

We will clarify that we are referring to the headwater stream sampling sites, which are paired across eight infrastructure categories.

R1 P4 L14-20: Some of this description of what types of infrastructure were built when might go better in the introduction. Or at least, you might want to introduce the concept of change in design through time in the introduction.

We will incorporate more background related to urban sanitary and stormwater infrastructure into the introduction and pare down our methods section accordingly.

R1 P4 L12-16: This sentence has a bit of a run-on feel; consider breaking down. Also, does "stormwater infrastructure... encompass older designs" *and* the newer GI ones? The way the sentence breaks doesn't suggest so. You could say, "We define stormwater infrastructure broadly to encompass older designs such as stormwater drainage networks and newer forms of 'green' stormwater infrastructure (GI)," and then define each in a sentence (or so) each.

Reviewer 1 is correct in his/her reading of this sentence. We will clarify our meaning here as the reviewer has recommended.

R1: P5 L20: Unclear how GIS calculations in previous sentence are used; abrupt transition back to "these surveys" is hard to follow.

We used the latitude and longitude of sampling sites to delineate watersheds and also calculate distance along the stream network for each location. We will clarify this in the text.

R1 P5 L25: "Relative contributions of inflow" *to groundwater*?

This sentence should say 'relative contributions of groundwater inflow to discharge at a given sampling point.' However, upon reflection and based on the reviewer's comments, we will remove this hydrologic mass-balance analysis from the paper since it is not directly discussed or used to interpret GHG results.

R1 P12 L30 - P13 L1: Consider referencing figures here (and more elsewhere in the discussion) to make it easy for readers to look back at the ratios etc. that you mention.

We will add a citation for figure 4 here.

R1 P13 L27 -P14 L9 & P15 L5-9: Most of this information should go in the introduction. You can refer back to it here insofar as your findings update or add to it, but it's unclear that they do. It does not seem entirely relevant here.

We will move this background information about N loading to urban streams in Baltimore to the introduction.

R1 P14 L31-32: You do not make it clear how this information about plants is relevant. Are you saying that some other type of plant within the waters you surveyed might be releasing methane in this way, but you didn't measure it? There are no transitions into or out of this part about the plants, either.

We meant here to provide some potential explanation for why streams in Wilcok and Sorrell (2008) found such high CH_4 emissions compared with our streams. We can shorten this section, to simply say that their methane emissions are not quite comparable because they included measurements of fluxes from aquatic plants, which would skew the comparison. To add more detail, we could include that these aquatic plants increase methane fluxes from sediment to atmosphere because of holes in their stems (aerenchyma), which allow for diffusive gas exchange between the atmosphere and rooting zone.

R1 P15 (26-27): It is unclear how exactly this part about wastewater relates to your results. Either make your transitions more clear, or move this sentence to a different section.

We will re-frame these concluding paragraphs to clarify the transitions here as follows. We present evidence in this paper that N from septic plumes and sewer lines is the principal source of N_2O saturation in our study sties. Dissolved inorganic N is highly correlated with N_2O in our study sites, and the highest values are only present in

watersheds with aging sewer infrastructure or septic systems. Our observations of N_2O saturation and emissions from urban and suburban headwater streams are some of the highest reported in the literature, comparable with streams and ditches in intensive agricultural watersheds (Harrison and Matson. 2003; Outram et al. 2012). These results suggest that streams draining low-density suburban or exurban land cover may be comparable to those in intensively fertilized agricultural areas in terms of N_2O emissions, however further study is necessary to constrain emission factors in non-agricultural landscapes.

R1 P15(28): You have not brought up the concept of mitigation before, and it isn't immediately obvious if mitigation per se is the goal, or how your results translate to doing mitigation. Elaborate.

We will remove the mention of mitigation since this is not the focus of this study, however we will add emphasis on the importance of accounting for N_2O urban streams (see our response to the previous comment).

R2 Comment 4) The dynamics of CO_2 are not considered in the discussion section

We did not focus on the CO_2 results in the discussion for two reasons. Firstly, CO_2 was strongly correlated with N_2O (as mentioned in the abstract and elsewhere), so additional descriptions of the spatial and temporal patterns seemed redundant. Secondly, as Reviewer 2 points out, we do not have the data to take into account abiotic sources of CO_2 and are therefore cautious to compare absolute values across systems.

R2 Comment 5) Reference to relevant recent studies on GHG dynamics in urban streams are missing (e.g. see Alshboul et al. 2016 Environmental Science & Technology 50: 5555-5563 DOI: 10.1021/acs.est.5b04923 and references therein).

We will incorporate this reference and papers cited therein it into the section of our discussion linking wastewater to patterns in aquatic GHGs.

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