ATMOSPHERIC METHANE EMISSIONS FOR ARGENTINA. COMPARISON WITH TROPOMI SATELLITE MEASUREMENTS.

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ABSTRACT:

Methane emissions have very important effect on global radiative forcing. Therefore, reducing these emissions has been proposed as an effective short-term strategy to mitigate global warming, in parallel with reductions in long-lived carbon dioxide (CO₂) for longterm temperature stabilizations. In this context, Argentina emits 3645 Gg of CH₄ mainly from livestock production, biomass burning and natural gas production. Since 2018, TROPOMI instruments provide global coverage on methane column-average mole fraction of dry air (XCH₄), and height profiles of methane concentrations. We compare two available methane inventory: a national (a high resolution of own ellaboration: GEAA) and an international (EDGAR) emissions database with TROPOMI measurements. By performing inverse satellite retrieval we evaluate the ability of remote sensing information to detect possible hotspot methane emissions and compare these results with the two inventories. From these analyzes, we observe that the latitudinal averages of the continental sector increase at a rate of 10 ppb/degree, from south to north, while the maritime sector remains constant. From a temporary perspective, the average monthly concentration amplitude range varies 40 to 50 ppb, with minimum values in March and maximum values in September.

1. INTRODUCTION

Short-lived climate pollutants (SLCP) including methane (CH4), black carbon (BC), tropospheric ozone, and hydrofluorocarbons (HFCs), have very important effect on global radiative forcing (Shindell et al., 2004; Etminan et al., 2016). Reducing these emissions has been proposed as an effective short-term strategy to mitigate global warming, together with reductions in long-lived carbon dioxide (CO₂) for long-term temperature stabilizations. Without reductions in both CO₂ and SLCPs, temperature increases are likely to exceed 1.5°C during the 2030s and exceed 2°C by mid-century (Shindell et al, 2017; Shoemaker et al., 2013, Collins et al, 2013, Ramanathan and Xu, 2010). On the other hand, methane is also a precursor to the formation of surface ozone, which affects population health (West et al., 2006; Isaken et al., 2009).

Atmospheric observations have shown significant increases in methane levels during the 20th century at a constant rate of approximately 15 ppb/year, with some stabilization in the 1990s and growing rapidly after 2006 (CDIAC, 2019) Blake, 2013; IPCC 2007, Fowler at al., 2009). The causes of these slope changes are not well known, but they do show a complex feedback of methane and biosphere. This important feedback and uncertainties about methane sources and sinks have motivated many researchers to conduct more detailed methane inventories and analysis.

Argentina emits 3200 Gg of CH₄ mainly from livestock production, biomass burning and natural gas production. Since 2018, TROPOMI instruments provide global coverage on methane column-average mole fraction of dry air (XCH₄), and height profiles of methane concentrations. We compare in this paper, two available methane inventory: a national (high

2. INVENTORIES

2.1 National methane inventories

In South America, there are national greenhouse gases (GHG) inventories submitted to the Intergovernmental Panel on Climate Change (IPCC) with a spatial resolution of provinces or districts. For Argentina, a national emissions database has been published for GHGs (TNCA, 2016). Air pollutants inventories, including methane, have been compiled for several sectors (GEAA inventory) at a high resolution $0.025^{\circ} \times 0.025^{\circ}$ (Puliafito et al., 2015, 2017, 2019). Castesana et al., (2018) presented a NH₃ inventory of agricultural activities with spatial resolution at the district level.

2.2 EDGAR emissions inventory

The EDGAR inventory compiles information from emission sources globally at a 0.1° (lat./long.) resolution. Methane emissions for all sources were compared with the GEAA inventory (Table 1).

resolution of own ellaboration, here called "GEAA") and an international Emissions Database for Global Atmospheric Research (EDGAR) (Janssens-Maenhout et al., 2017) emissions database: EDGARv4.2FT2010; with the TROPOspheric Monitoring Instrument (TROPOMI), on board the Copernicus Sentinel-5 Precursor satellite, (TROPOMI, 2018).

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Figure 1. Methane inventory: a) EDGAR, b) GEAA

Figure 1 shows two methane inventories (EDAGR and GEAA). Table 1 shows total emissions by sector for Argentina, while Figure 2 show a series for the three mentioned inventories for years 1990-2016. Notice that EDGAR is only available up to 2012, while TNCA was calculated until 2014 and GEAA is available to 2016.

Comparing sectoral methane emissions, from enteric fermentation, manure management, biomass burning and rice

cultivation, for years 1990-2012; EDGAR averages 3155 ± 390 Gg/year; while GEAA averages 3030 ± 235 Gg/year. Thus, there is a 4% relative difference between both inventories. However, other discrepancies for CH₄ emissions concerning several subsectors were present (i.e. fugitive emissions from oil and gas: EDGAR 864 Gg, GEAA: 272 Gg), rising the differences for all sector (2012) to 1050 Gg (22%): EDGAR: 4557 Gg, GEAA: 3644 Gg. Despite most sectors, have good agreements, spatial distribution presented some differences.

Therefore, EDGAR overestimated on average 13% compared to GEAA and TNCA inventories for the considered period. Estimations of methane emissions corresponding to year 2012 for agriculture, livestock and biomass burning were 3257.46 Gg for EDGAR, 2454.40 for GEAA and 2505.56 Gg for TCNA.

Sector	GEAA	EDGAR	TNCA
Ref. Year	2014	2012	2014
Electricity	4.58	3.08	5.56
Transport	19.40	14.68	17.26
Residential +	4.17	10.89	4.32
Industries			
Fugitive emissions	272.13	864.67	327.23
Enteric	2,620.23	3,110.61	2,413.74
Fermentation			
Manure	29.14	59.86	41.65
management			
Rice cultivation	30.08	27.93	28.77
Biomass burning	39.20	59.06	93.73
Urban Waste	625.97	406.14	625.97
Total	3,644.90	4,556.91	3,558.25

Ref: adapted from Puliafito, et al (2017, 2019); (*) includes AWB: Agricultural waste burning. TNCA, (2016); EDGAR: Janssens-Maenhout et al., (2017).

Table 1: Total methane emissions (Gg/year) from all sectors for Argentina 2016.



Figure 2. Comparison of methane emissions for Argentina

Since GEAA has a higher spatial resolution $(0.025^{\circ} \text{ lat./long.})$, this inventory needed to be converted to a 0.1° resolution to allow an adequate spatial comparison. Therefore, each 0.1° broad cell was computed by adding all corresponding 0.025° higher-resolution cells within the circumscribed broader-resolution cell.

EDGAR presented higher values than GEAA in cities and in mountain range (where almost no activity is present). It is observed in Figure 1, that in Patagonia (Southern Argentina latitudes 40° S – 55° S and western border) EDGAR map show average values around 20 Mg/year, while GEAA show null values for the same regions. A second comparison included the geographical extent of EDGAR (Figure 1a) and GEAA (Figure

This contribution has been peer-reviewed. The double-blind peer-review was conducted on the basis of the full paper. https://doi.org/10.5194/isprs-annals-IV-3-W2-2020-107-2020 | © Authors 2020. CC BY 4.0 License. Primary publication at IEEE Xplore: https://doi.org/10.1109/LAGIRS48042.2020.9165602

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1b) inventories. Both inventories were also compared by totalizing 0.1° bin latitudinal and longitudinal methane emissions (Figure 3). Also high fugitive emissions from natural gas pipelines can be clearly observed in EDGAR map. EDGAR presented higher values than GEAA specially between 42° S -34° S and 69° W $- 66^{\circ}$ W. This area corresponds to a natural gas producing area, whose difference were associated mainly to venting from exploring and productive natural gas wells. This high difference is consistent with Table 1. In fact, years 2010-2016, have been very active but variable years with respect to oil and natural gas exploration and production, and therefore the differences may be explained by different activity data.



Figure 3. Latitudinal averages for EDGAR and GEAA methane inventory for 0.1° resolution.

3. SATELLITE DATA

3.1 SCHIAMACHY AND GOSAT

Global methane data has been published in the form of columnaveraged dry mole fraction XCH4 from SCIAMACHY (Scanning Imaging Absorption Spectrometer for Atmospheric Chartography, on board ENVISAT, ground resolution: 30 km × 60 km), years 2002-2009, (Frankenberg et al., 2005, 2011). and GOSAT (Greenhouse gases observing satellite, ground resolution: 10 km × 60 km), for years 2009-2018, (Yokota, 2004, Yoshida, 2011, Schepers et al., 2012).

Figure 4 shows monthly zonal mean (20°S-55°S) of methane column average dry air mixing ration XCH4 (ppb) from SIAMACHY and GOSAT (2003-2017). The annual averages have varied from 1716±36 ppb in 2003 to 1780±27 ppb in 2017. Figure 5 shows the monthly surface concentrations of methane measured at the GAW station (Global Atmospheric Watch Station, from the World Meteorological Organization, operated by the National Meteorological Service of Argentina) at the city of Ushuaia (54.8°S, 64.3°W)(GAW, 2018). Monthly average concentration values in Ushuaia had a seasonal amplitude of 40-50 ppb, with minimum values in March and maximum values in September with an average annual rate of increase of 0.35% (6.2 ppb/year) since 2008. Three slopes are identify: (1994-1999), (2000-2007) and (2008-2017), which agree with global tendencies. Although there are many uncertainties regarding the stable period (2000-2007), Kirschke et al. (2013) attribute it to a reduction-stabilization of global emissions and an increase in stabilization of microbial activity.







Figure 5. monthly surface concentrations of methane measured at the GAW station in Ushuaia (54.8°S, 64.3°W)(GAW, 2018).

3.2 TROPOMI measurements

The newly TROPOMI instruments provided information (since 2018) on methane column-average mole fraction of dry air (XCH4), and height profiles of methane concentrations, with ground resolution of 7 km \times 3.5 km. For TROPOMI XCH4 data, we used Level 2 product (Apituley et. al., 2017) over Argentina between May 2018 and April 2019 with a resolution of 0.25° \times 0.25°. TROPOMI data covered almost the entire spatial extension of Argentina with sufficient number of pixels.

Figure 6 shows annual average of XCH4 over Argentina using TROPOMI (May 2018- April 2019). We used TROPOMI data because of its higher spatial resolution and coverage (than SIAMACHY and GOSAT). It can be seen that methane concentrations increased from south to north (in the SE-NW direction) coinciding over the continental sector with an average rate of 2.2 ± 0.5 ppb/degree (N-S) and 1.4 ± 0.6 ppb/degree (W-E). To qualitativelly compare the methane inventory with satellite data we followed Jacob et al, (2016) (i.e. Eq. 13 and Table 2) inverse calculation using TROPOMI data. A mean methane enhancement Δ XCH₄ is estimated by subtracting a background to the satellite data.



Figure 6: Methane column average dry air mixing ration (ppm) from two satellites for year 2018. a) TROPOMI; b) Estimated background; c) methane enhancement Δ XCH₄ from satellite; d) methane enhancement Δ XCH₄ from inventory



This contribution has been peer-reviewed. The double-blind peer-review was conducted on the basis of the full paper. https://doi.org/10.5194/isprs-annals-IV-3-W2-2020-107-2020 | © Authors 2020. CC BY 4.0 License. Primary publication at IEEE Xplore: https://doi.org/10.1109/LAGIRS48042.2020.9165602 This background (Figure 6b) is calculated as a plane (slope= 0.09 ppb/°Lat, initial value = 1760 ppb @ 56S,75W), which increasead from south to north. This values are consistent with slopes calculated from SCHIAMACHY and GOSAT (Figure 4). The retrieved emission map (Figure 6c) was estimated using Eq. 13 from Jacob et al., (2016) (W=100 km, pressure map from TROPOMI, and u=23), which should be compared to Figure 6d. This is calculated as a smoothed map of Figure 1b, using a gaussian convolution of 0.3°. Figure 6c, gives a qualitative insight into the methane emissions inventory, capturing the main area of emissions: livestock production and agriculture in the central area (38-32S, 40-45W) and biomass burning at North East areas (27-25S, 60-57W). It can be seen, that the GEAA inventory understimated the CH4 emissions in the NE area. Probably the retrieval calculation is including methane transport from biomass burning from other areas (i.e Amazonia), as it is shown in Figure 6a, but not included in the present inventory.

4. DISCUSSION

Argentina is an important agriculture and livestock producer, reaching 123 million tons of crops annually and breeding 52 million heads of cattle, emitting in year 2016, 2186.10 Gg methane from the beef cattle sector, 288.40 Gg from dairy cattle sector and another 204.58 Gg from other livestock production, totaling 2679.08 Gg. This emission represents 83% of the 3198.45 Gg total national methane emissions (all sectors) and 27% of total national GHG emissions. Other sectors as the energy-producing sector (electricity and fugitive form oil/natural gas extraction) added 278 Gg of methane.

A high-resolution methane emissions inventory for Argentina (GEAA) was compared to an international database (EDGAR) and The Third National Greenhouse Report (TNCA, 2016). There are two main differences, however, with the two abovementioned inventories: a) GEAA inventory includes species that affect air quality and b) it display them on a higher spatial resolution map (0.025° resolution). Compared to EDGAR international emissions database (0.1° spatial resolution) the methane emissions for Argentina for all sector gave an average difference of 13% and a spatial uncertainty of 50 Mg/year/cell.

The present study also included the analysis SCIAMACHY, GOSAT AND TROPOMI satellite data for the analysis of methane concentrations in the Argentine territory. The aim of this analysis was to explore if methane emissions could be detected from inverse modelling from satellite sensors.

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REFERENCES

Apituley, A., Pedergnana, M., Sneep, M., Pepijn Veefkind, J., Loyola, D., Hasekamp, O., 2017: Sentinel-5 precursor/TROPOMI Level 2 Product User Manual Methane, SRON-S5P-LEV2-MA-001,

http://www.tropomi.eu/sites/default/files/files/Sentinel-5P-

Level-2-Product-User-Manual-Methane.pdf, Last accesed, Sept. 2019.

Blake. D., 2013. Methane, Nonmethane Hydrocarbons, Alkyl Nitrates, and Chlorinated Carbon Compounds including 3 Chlorofluorocarbons (CFC-11. CFC-12. and CFC-113) in Whole-air Samples. Carbon Dioxide Information Analysis Center. Oak Ridge National Laboratory. U.S. Department of Energy. Oak Ridge Tenn.. U.S.A. http://cdiac.ess-dive.lbl.gov/trends/otheratg/blake/blake.html

Butz A., Guerlet S., Hasekamp O., Schepers D., Galli A., Aben I., Frankenberg C., Hartmann J., Tran H., Kuze A., Keppel-Aleks G., Toon G., Wunch D., Wennberg P.,Deutscher N., Griffith D., Macatangay R., Messerschmidt J., Notholt J. and Warneke T., 2011. Toward accurate CO2 and CO4 observations from GOSAT, *Geophys. Res. Lett.*, 38, L14812, https://doi:10.1029/2011GL047888

Castesana, P., Dawidowski, L., Finster, L., Gómez, D., Taboada, M., 2018. Ammonia emissions from the agriculture sector in Argentina; 2000–2012, *Atmospheric Environment* 178, 293-304. https://doi.org/10.1016/j.atmosenv.2018.02.003 CDIAC (2019): NOAA-CDIAC data http://cdiac.essdive.lbl.gov/methane.html

Collins, M., R. Knutti, J. Arblaster, J.-L. Dufresne, T. Fichefet, P. Friedlingstein, X. Gao, W.J. Gutowski, T. Johns, G. Krinner, M. Shongwe, C. Tebaldi, A.J. Weaver, and M. Wehner, 2013: Long-term climate change: Projections, commitments and irreversibility. In Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. T.F. Stocker, D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Doschung, A. Nauels, Y. Xia, V. Bex, and P.M. Midgley, Eds. Cambridge University Press, pp. 1029-1136, doi:10.1017/CB09781107415324.024.

Etminan M., G. Myhre, E. J. Highwood and K. P. Shine., 2016. Radiative forcing of carbon dioxide methane and nitrous oxide: A significant revision of the methane radiative forcing. *Geophys. Res. Lett.* 43. 12.614–12.623. https://doi:10.1002/2016GL071930

Fowler D.; Pilegaard. K.; Sutton. M.A.; Ambus. P.; Raivonen. M.; Duyzer. J.; Simpson. D.; Fagerli. H.; Fuzzi. S.; Schjoerring. J.K.; et al., 2009. Atmospheric composition change: Ecosystems-Atmosphere interactions. *Atmos. Environ.* 43.5193–5267. https://doi.org/10.1016/j.atmosenv.2009.07.068

Frankenberg C., Aben I., Bergamaschi P., Dlugokencky E., van Hees R., Houweling S., van der Meer P., Snel R., and P. Tol., 2011. Global column-averaged methane mixing ratios from 2003 to 2009 asderived from SCIAMACHY: Trends and variability, *J. Geophys. Res.*, 116, D04302, https://doi:10.1029/2010JD014849.

Frankenberg C., Meirink J., van Weele M., Platt U., and Wagner T., 2005. Assessing methane emissions from global space-borne observations, *Science*, 308, 1010–1014. https://doi:10.1126/science.1106644

GAW-Global Atmospheric Watch, 2018. A program from World Meteorological Organization. http://www.wmo.int/pages/prog/arep/gaw/ghg/ghgbull06_en.ht

This contribution has been peer-reviewed. The double-blind peer-review was conducted on the basis of the full paper. https://doi.org/10.5194/isprs-annals-IV-3-W2-2020-107-2020 | © Authors 2020. CC BY 4.0 License. Primary publication at IEEE Xplore: https://doi.org/10.1109/LAGIRS48042.2020.9165602 ml and World Data Center for Greenhouse Gases (WDCGG) https://gaw.kishou.jp/

IPCC - The Intergovernmental Panel on Climate Change. 2007. The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment. Report of the Intergovernmental Panel on Climate Change; Solomon S., Qin D., Manning M., Chen Z., Marquis M., Averyt K.B., Tignor M., Miller H.L, Eds.; Cambridge University Press: Cambridge. UK/New York. NY. USA.

Isaksen I., Granier. C., Myhre. G., Berntsen T.K., Dalsøren S.B., Gauss M., Klimont Z., Benestad R., Bousquet P., Collins W., Cox T., Eyring V., Fowler. D., Fuzzi S., Jöckel P., Laj. P., Lohmann U., Maione M., Monks P., Prevot A., Raes F., Richter A., Rognerud B., Schulz M., Shindell D., Stevenson D., Storelvmo T., Wang W.-C., 2009. Atmospheric composition change: Climate-Chemistry interactions, *Atmospheric Environment*, 43, (33), pp. 5138-5192. https://10.1016/j.atmosenv.2009.08.003

Jacob, D., Turner, A., Maasakkers, J., Sheng, J., Sun, K., Liu, X., Chance, K., Aben, I., McKeever, J., and Frankenberg, C. Satellite observations of atmospheric methane and their value for quantifying methane emissions. *Atmos. Chem. Phys.*, 16, 14371–14396, 2016, doi:10.5194/acp-16-14371-2016

Janssens- Maenhout G., Crippa M., Guizzardi D., Muntean M., Schaaf E., Dentener F., Bergamaschi P., Pagliari V., Olivier J., Peters J., van Aardenne J., Monni S., Doering U., Petrescu A., 2017. EDGARv4.3.2 Global Atlas of the three major Greenhouse Gas Emissions for the period 1970-2012. *Earth Syst. Sci. Data Discuss.* https://doi.org/10.5194/essd-2017-79.

Puliafito, S. E.; Allende, D., Castesana, P., Ruggeri, M., 2017. High-resolution atmospheric emission inventory of the argentine energy sector. Comparison with Edgar global emission database. *Heliyon* 3 e00489. https://doi.org/10.1016/j.heliyon.2017.e00489

Puliafito, S.E., Allende, D., Pinto, S., Castesana, P., 2015. High resolution inventory of GHG emissions of the road transport sector in Argentina, *Atmospheric Environment* 101, 303-311, https://doi.org/10.1016/j.atmosenv.2014.11.040

Puliafito, S. E., Bolaño-Ortiz, T. Berná, L., Pascual-Flores, R., 2019. High-resolution inventory of atmospheric emissions from livestock production, agriculture, and biomass burning sectors of Argentina, *Atmospheric Environment*, https://doi.org/10.1016/j.atmosenv.2019.117248

Ramanathan, V. and Xu, Y., 2010, The Copenhagen Accord for limiting global warming: Criteria, constraints, and available avenues, *PNAS*, 107 (18) 8055-8062; https://doi.org/10.1073/pnas.1002293107

Schepers D., Guerlet S., Butz A., Landgraf J., Frankenberg C., Hasekamp O., Blavier J., Deutscher N., Griffith D., Hase F., Kyro E., Morino I., Sherlock V., Sussmann R. and I. Aben I., 2012. Methane retrievals from Greenhouse Gases Observing Satellite (GOSAT) shortwave infrared measurements: Performance comparison of proxy and physics retrieval algorithms, *J. Geophys. Res.*, 117, D10307, https://doi:10.1029/2012JD017549 Shindell, D., Borgford-Parnell, N., Brauer, M., Haines, A., Kuylenstierna, J., Leonard, S., Ramanathan, V., Ravishankara, A., Amann, M., Srivastava, L., 2017: A climate policy pathway for near- and long-term benefits, *Science* 356 (6337): 493-494, May 2017, doi: 10.1126/science.aak9521

Shindell. D.T.. Walter. B.P.. Faluvegi. G., 2004. Impacts of climate change on methane emissions from wetlands. *Geophysical Research Letters*, 31. L21202. https://doi:10.1029/2004GL021009

Shoemaker, J., Schrag, D., Molina, M., Ramanathan, V., 2013, What Role for Short-Lived Climate Pollutants in Mitigation Policy? *Science* 342 (6164), 1323-1324, Dec. 2013, doi: 10.1126/science.1240162

TCNA (2016) Argentina Third National Greenhouse Report to UNFCCC., 2016. www.ambiente.gob.ar/tercera-comunicacion-nacional/

TROPOMI (2018): TROPOspheric Monitoring Instrument: http://www.tropomi.nl/?lang=en, last Access October 2019).

West I.J., Fiore A., Horowitz L., Mauzerall D., 2006. Global health benefits of mitigating ozone pollution with methane emission controls *PNAS*. March 14. vol. 103. no. 11. 3988–3993. www.pnas.orgcgidoi10.1073pnas.0600201103

Yokota T., Oguma H., Morino I., Higurashi A., Aoki T., and G. Inoue., 2004. Test measurements by a BBM of the nadirlooking SWIR FTS aboard GOSAT to monitor CO2 column density from space, *Proc. SPIE Int. Soc. Opt. Eng.*, 5652, 182.

Yoshida, Y., Y. Ota, N. Eguchi, N. Kikuchi, K. Nobuta, H. Tran, I. Morino, and T. Yokota., 2011. Retrieval algorithm for CO2 and CO4 column abundances from short-wavelength infrared spectral observations by the greenhouse gases observing satellite, *Atmos. Meas. Tech.*, 4, 717–734; https://doi.org/10.5194/amt-4-717-2011