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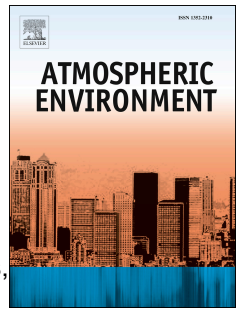

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Wet nitrogen (N) deposition to urban Latin America: filling in the gaps with GEOS-Chem

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Abstract

In Latin America, atmospheric deposition is a major vector of nitrogen (N) input to urban systems. Yet, measurements of N deposition are sparse, precluding analysis of spatial patterns, temporal trends, and ecosystem impacts. Chemical transport models can be used to fill these gaps in the absence of dense measurements. Here, we evaluate the performance of a global 3-D chemical transport model in simulating spatial and interannual variation in wet inorganic N (NH_4-N + NO_3-N) deposition across urban areas in Latin America. Monthly wet and dry inorganic N deposition to Latin America were simulated for the period 2006-2010 using the GEOS-Chem Chemical Transport Model. Published estimates of observed wet or bulk inorganic N deposition measured between 2006-2010 were compiled for 16 urban areas and then compared with model output from GEOS-Chem. Observed mean annual inorganic N deposition to the urban study sites ranged from 5.7-14.2 kg ha^{-1} yr^{-1}, with NH_4-N comprising 48-90% of the total. Results show that simulated N deposition was highly correlated with observed N deposition across sites (R^2 = 0.83, NMB = -50%). However, GEOS-Chem generally underestimated N deposition to urban areas in Latin America compared to observations. Underestimation due to bulk sampler dry deposition artifacts was considered and improved bias without improving correlation. In contrast to spatial variation, the model did not capture year-to-year variation well. Discrepancies between modeled and observed values exist, in part, because of uncertainties in Latin American N emissions inventories. Our findings indicate that even at coarse spatial resolution, GEOS-Chem can be used to simulate N deposition to urban Latin America, improving understanding of regional deposition patterns and potential ecological effects.

Keywords: air pollution, chemical transport models, cities, critical loads, livestock, rainwater chemistry

1. Introduction

In Latin America, atmospheric deposition is a major, and sometimes the primary, vector of nitrogen (N) input to terrestrial ecosystems (Austin et al. 2013; Schwede et al. 2018). Nitrogen is a critical limiting nutrient in many ecosystems and a harmful pollutant when supplied in excess of plant requirements (Vitousek and Howarth 1991; Erisman et al. 2013). Between 1961-2009, anthropogenic N inputs to Latin America from atmospheric deposition, fertilizer use, N fixation, and imports and exports of N in agricultural commodities (i.e., food and feed) increased from 1.94 to 7.91 kg N ha^{-1} yr^{-1} (Han et al. 2020). In 2009, atmospheric wet plus dry oxidized N deposition comprised ~16% of this total (Han et al. 2020). Although estimates remain uncertain for many regions in Latin America, global models indicate values of wet N deposition to South America ranging from 5-15 kg ha^{-1} yr^{-1}, with deposition dominated by reduced N (Aas et al. 2014).

Over the next several decades, rising N emissions from urbanization, biomass burning, and agricultural expansion and intensification will contribute to further increases in N deposition across the region (Martinelli et al. 2006; Lamarque et al. 2013; Galloway et al. 2021). Agriculture is a key economic sector in Latin America, with crop and livestock production representing major and growing sources of NH_3 and N_2O to the atmosphere (Austin et al. 2006; Bustamante et al. 2014; Steinfeld and Wassenaar 2007). By some estimates, fertilizer N emissions will be on par with those in China by 2050 (Alexandratos and Bruinsma 2012). Biomass burning emissions exhibit considerable spatiotemporal variability across South America (Castellanos et al. 2014) but remain an important contributor to total NO_x emissions (Jaeglé et al. 2005) and N deposition near fire-prone regions (Chen et al. 2010). Fossil fuel combustion represents another potentially large source of atmospheric N in rapidly expanding urban and industrial areas (Filoso et al. 2006).

Notwithstanding rising N emissions, N deposition to and impacts on Latin American ecosystems are infrequently measured and monitored (e.g., Aas et al. 2014; Carnelos et al. 2019; Ometto et al. 2020). Compared to North America, Europe, and Asia, vast land areas in Latin America remain under-sampled for N inputs (Ponette-González et al. 2014; Vet et al. 2014). Formal networks to monitor atmospheric wet deposition are sparse and dry deposition measurements are limited (Ometto et al. 2020). Thus, existing measurements of N deposition preclude assessment of long-term temporal trends and characterization of spatial patterns based

on observed data (Weathers and Ponette-González 2011; Vet et al. 2014; Carnelos et al. 2019). Global and regional atmospheric chemistry transport models can be used to fill these gaps (Dentener et al. 2006; Lamarque et al. 2013; Vet et al. 2014; Schwede et al. 2018; Ackerman et al. 2019), but in Latin America extensive ground-based measurements for model evaluation are lacking. This is especially true for urban areas (Holland et al. 2005), where models are also poorly constrained by the dearth of flux studies needed for modeling of dry deposition and difficulties associated with building complex urban surfaces into model structure (Ching 2013).

In Latin America, >80% of the population resides in urban areas (UNPD 2018). A major fraction of this population (6-34% depending on the country) is concentrated in megacities (population >10 M), including Mexico City, Bogotá, Lima, São Paulo, Rio de Janeiro, and Buenos Aires (UNPD 2018). Megacities are often the most polluted cities in Latin America, but air quality problems also plague mid-sized cities (Jorquera et al. 2019). As such, air quality monitoring is widespread in urban areas (Jorquera et al. 2019), but there remain few corresponding measurements of atmospheric wet or dry N deposition (Decina et al. 2020).

Improved estimates of N deposition to Latin America urban areas are needed to better understand regional deposition patterns and potential ecological effects (Phoenix et al. 2006; Ometto et al. 2020). Tropical ecosystems vulnerable to elevated N, for example N-rich tropical moist forests, encompass nearly half of Latin America's land cover (Aide et al. 2013). These ecosystems are often situated in and downwind of urban deposition 'hotspots' (Decina et al. 2020), where annual N deposition can exceed 5-10 kg ha^{-1} yr^{-1} (e.g., Fenn et al. 1999; Ponette-González et al. 2010; de Souza et al. 2015), the critical load limit for plant community composition and nitrate (NO_3^-) leaching in tropical humid forest (Pardo et al. 2011). Indeed, elevated N deposition to urban and near-urban tropical forest has been found to enhance soil N availability (Cusack 2013; Ponette-González et al. 2017), increasing the potential for NO_3^- leaching losses to surface and groundwater (e.g., Fenn et al. 1999). Increased gaseous N fluxes to the atmosphere may also be expected in N-polluted urban areas (Fang et al. 2015). Rising N deposition may thus degrade air and water quality and contribute to shifts in plant community composition, effects that can extend tens of kilometers downwind (Du et al. 2015).

In this study, our objective was to evaluate the performance of a global 3-D chemical transport model in simulating spatial and interannual variation in wet inorganic N (NH_4-N + NO_3-N) deposition across 16 urban areas (pop ~0.1 – 20 M) in Latin America. Although we here

focus on wet deposition due to the lack of ground-based dry deposition measurements in the study areas, we note that dry deposition can comprise a significant fraction of total (wet + dry) atmospheric deposition to ecosystems (Weathers et al. 2006; Schwede and Lear 2014). We build on previous model evaluations by focusing on the period 2006-2010.

2. Material and methods

2.1 GEOS-Chem simulated estimates of atmospheric N deposition

We simulated monthly inorganic N in wet (precipitation) and dry (gases and particles) deposition to Latin America using the GEOS-Chem Chemical Transport Model (v8-03-02; www.geos-chem.org) (Figure 1). GEOS-Chem was run for the period 2006-2010 at 2° x 2.5° horizontal resolution. Simulated wet inorganic N deposition included the chemical species ammonium (NH_4), ammonia (NH_3), nitrate (NO_3), and nitric acid (HNO_3), and dry N deposition included the chemical species NH_3, particulate NH_4, HNO_3, nitrogen dioxide (NO_2), peroxyacyl nitrate species (PAN, PMN, PPN), organic nitrates (R_4N_2), NO_2+NO_3 adduct (N_2O_5), and particulate NO_3.

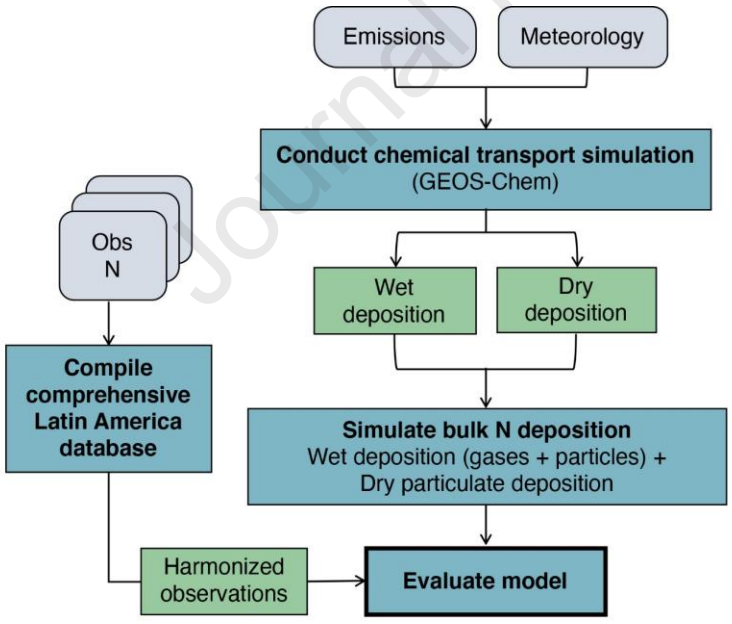

Figure 1. Simplified flow chart of the inputs (grey curved boxes), outputs (green rectangles), and processes (blue rectangles) used in the evaluation of GEOS-Chem for simulating atmospheric wet inorganic N deposition to urban areas in Latin America.

GEOS-Chem, described in detail by Bey et al. (2001), uses global emission inventories superseded by regional inventories where available. Emissions are separated into four categories: anthropogenic, biofuel, biomass, and biogenic. With the exception of Mexico, where the model uses the Big Bend Regional Aerosol & Visibility Observational Study (BRAVO) emission inventory (base year 1999), Latin American countries use anthropogenic emissions from the Emission Database for Global Atmospheric Research (EDGAR, https://edgar.jrc.ec.europa.eu). Anthropogenic emissions from EDGAR as implemented in GEOS-Chem v8-03-02 include those associated with transportation, industrial, residential, and traditional fossil fuel energy production, as well as biofuel, biomass, shipping, aircraft, and fertilizer emissions. Nitrogen oxides from fertilized soils and aircraft are included globally. Shipping emissions are represented by the global International Comprehensive Ocean-Atmosphere Data Set (https://icoads.noaa.gov), and fire emissions are represented with the monthly average Global Fire Emission Database (GFED2, https://globalfiredata.org). For natural biogenic emissions, we use GEOS-Chem's online implementation of the Model of Emissions of Gases and Aerosols from Nature (https://bai.ess.uci.edu/megan). Lightning emissions were enabled and climatologically redistributed using the OTD approach. EDGAR was used for all species except NH_3, which is supplied by the Global Emissions InitiAtive (GEIA, Bouwman et al. 1997).

In this study, we used the Goddard Earth Observing System (GEOS) version 5 meteorological inputs (GEOS-5; Rienecker et al. 2008). In GEOS-Chem, advection was solved using the recommended monotonic piecewise parabolic method in both horizontal and vertical dimensions (Colella et al. 1984) with a 15-minute timestep (recommended for 2° x 2.5° resolution). A stratospheric boundary condition was used for ozone and NO_y. We also enabled cloud convection as well as planetary boundary layer mixing both with a timestep set to 15 min.

Wet deposition included particles and soluble gases in rain, snow, and sleet removed by in-cloud (rainout) and below-cloud (washout) processes. In GEOS-Chem, wet deposition includes scavenging in convective updrafts and large-scale cloud systems (Liu et al. 2001; Amos et al. 2012). Dry deposition included fluxes of N in the form of particles and gases. Gaseous dry deposition was simulated with a big-leaf resistance-in-series model (Zhang et al. 2012). Bidirectional flux of NH_3 was not considered in the model, however we assumed a net downward flux in urban areas. Wet and dry deposition were calculated by summing species-

specific deposition values. GEOS-Chem output was converted to kg N ha^{-1} mo^{-1} for comparability with observations.

2.2 Observations of N deposition

We used Google Scholar and Web of Science to search for peer-reviewed publications in English and Spanish that included wet or bulk (hereafter "observed") inorganic N deposition to urban areas in Latin America, from which we extracted data for analysis. Publicly available data were downloaded from Mexico City's Red de Depósito Atmosférico (REDDA; www.aire.cdmx.gob.mx), while for sites in Argentina and Uruguay data were obtained from RP-RainNet, a network located in the Rio de la Plata region (Carnelos et al. 2019). We also obtained data from ongoing studies where available.

Published estimates of wet-only (measured with an automated sampler which opens during and closes after precipitation events, e.g., National Atmospheric Deposition Program (NADP) National Trends Network) or bulk (measured with a sampler that remains open during collection periods, e.g., Ponette-González et al. 2017) inorganic N deposition measured between 2006-2010 were assembled for 16 urban areas in six countries in North, Central, and South America: Argentina, Brazil, Colombia, Costa Rica, Mexico, and Uruguay (Table 1). Because dry particulate N inputs to bulk collectors are small (Izquierdo and Avila 2013), we considered wet and bulk deposition to be comparable for the purposes of this study. Mean annual precipitation for these sites ranged from ~600-2000 mm yr^{-1}. A site was classified as "urban" if defined as such by the authors in the original publication; all urban sites were built-up areas with >2,500 inhabitants. Site IDs are provided in Table 1 and are used in the following sections.

Protocols for sampling precipitation varied among the studies. Bulk collectors were deployed at the majority (n=11) of the sites, while use of wet-only collectors was less common. In a few cases, multiple locations within a single large metropolitan area were sampled (i.e., San José, Mexico City), providing better geographic coverage of city-wide N deposition. Across studies, measurement periods ranged from <1 year to five years. Five sites sampled precipitation during the wet season only (i.e., BrIl, BrIt, CoMa, CoSa, and MeMe), and five sites included in the dataset are part of atmospheric deposition monitoring networks with ongoing collection of precipitation samples.

Volume-weighted mean concentrations (VWM) and deposition of NH_4-N and NO_3-N were recorded for each site. If N deposition was not reported, then deposition was calculated by multiplying VWM concentrations by precipitation amount. Rainfall data were obtained directly from the authors or network, downloaded from the nearest meteorological station to the site, or extracted from global gridded precipitation datasets. When multiple locations within a single urban area were sampled, the mean VWM reported by the authors was included in the dataset for this study as a single data point. When the authors did not include a mean VWM, the mean VWM for the urban area was calculated from the raw data. The temporal resolution of the data varied among the studies, from monthly to annual (Table 1). Because of this variation, N deposition was calculated per period (i.e., month, bimonth, season, year) for each year in the study (2006-2010) for comparison with model output.

203 **Table 1:** Location and characteristics of precipitation sampling protocol for the 16 Latin American urban areas included this analysis.

City	Site ID	Latitude	Longitude	Type	Sites	Measurement Period	Sampling Frequency	Temporal Resolution	Data Source
Pergamino	ArPe	-33.8836	-60.5669	Bulk	1	Dec 2006-Dec 2010	Monthly	Monthly	Carnelos et al. (2019)
Buenos Aires	ArBu	-34.5997	-58.3819	Bulk	1	Jan 2006-Dec 2010	Monthly	Monthly	Carnelos et al. (2019)
Ilhéus	BrIl	-14.7935	-39.046	Bulk	1	Sep 2009-Jan 2010 (Wet season)	Weekly	Seasonal	Araujo et al. (2015)
Itabuna	BrIt	-14.7880	-39.2784	Bulk	1	Sep 2009-Jan 2010 (Wet season)	Weekly	Seasonal	Araujo et al. (2015)
Rio de Janeiro	BrRi	-22.8967	-43.1322	Bulk	1	Aug 2008-Jul 2009	Weekly	Annual	de Souza et al. (2015)
Cubatão	BrCb	-23.85	-46.4166	Wet	1	Jun 2009-Aug 2010	Event	Annual	Vieira-Filho et al. (2015)
São Paulo	BrSa	-23.53	-45.65	Wet	2	Nov 2004-Oct 2006	Event	Annual	Vieira-Filho et al. (2010)
Porto Alegre	BrPo	-29.919	-51.1821	Wet	3	Jul 2005-Dec 2007	Event	Annual	Migliavacca et al. (2012)
Cuiabá	BrCu	-15.611	-56.0258	Bulk	1	Feb 2006-Nov 2009	Event	Annual	Marques et al. (2011)
Manizales	CoMa[a]	5.0661	-75.475	Bulk	4	Oct 2010-Apr 2011 (Wet season)	Event	Seasonal	González & Aristizábal (2012)
San José	CoSa1	9.9356	-84.0714	Bulk	11	Aug 2007-Nov 2007 (Wet season)	Event	Seasonal	Herrera et al. (2009)
Monterrey	MeMo	25.7247	-100.3154	Wet	1	Jan 2007-Dec 2007	Event	Annual	Rámirez Lara et al. (2010)
Xalapa	MeXa[a]	19.5241	-96.9392	Bulk	2	Jan 2006-Nov 2007	6-10 wks	Bimonthly	Ponette-González et al. (2010)
Mexico City	MeMe	19.4291	-99.1319	Wet	16	May-Oct 2006-2010 (Wet season)	Weekly	Monthly	REDDA

Montevideo	UrMo	-34.8669	-56.1666	Bulk	1	Oct 2006-Dec 2010	Monthly	Monthly	Carnelos et al. (2019)
La Paloma	UrPa	-34.6530	-54.1701	Bulk	1	Sep 2006-Dec 2010	Monthly	Monthly	Carnelos et al. (2019)

[a]Ammonium was not measured in this study.

205 **2.3 Model performance**

206 We evaluated the performance of GEOS-Chem for simulating inorganic N deposition to
207 Latin American urban areas using linear regression analysis, the normalized mean bias (NMB),
208 and spatial assessments. First, we used linear regression to compare model-simulated and
209 observed estimates, with R^2 values indicating how well the variability in the observed values was
210 predicted by the model. For the comparisons, GEOS-Chem monthly deposition values were
211 aggregated to match observational periods (i.e., month, bimonth, season, year).

212 The following regressions were performed for all sites and years: (1) GEOS-Chem
213 simulated wet N against observed N deposition; and (2) GEOS-Chem simulated wet + dry
214 particulate N against observed N deposition. Simulated wet plus dry particulate deposition,
215 which excluded all gaseous N species, was calculated because bulk collectors were used to
216 measure N concentrations in rainfall at most of the sites. It is generally thought that dry N
217 deposition to bulk collectors is minimal (Cook et al. 2018). However, we assumed that in
218 addition to wet deposition, some particulate N may have deposited into these bulk collectors
219 (Dämmgen et al. 2005; Izquierdo and Avila 2013). Second, we calculated the normalized mean
220 bias between the model and the observations in our dataset. The latter statistical measure was
221 used to compare the degree of model over- or underestimation across sites with a wide range of
222 wet N deposition values. Third, we compared spatial patterns of simulated and observed N
223 deposition. The spatial maps of simulated N represent mean annual N deposition for the period
224 2006-2010. Monthly simulated values were summed to produce annual values, and the annual
225 sums were then averaged to obtain mean annual N deposition in kg ha^{-1} yr^{-1}. Observed mean
226 annual N deposition was calculated in the same way. Deposition rates for sites (i.e., BrIl, BrIt,
227 CoMa, CoSa, MeMe) with wet season measurements were multiplied by a factor of 2-3 solely to
228 enable them to be plotted on the same scale.

229

230 **3. Results**

231 **3.1 GEOS-Chem model performance**

232 The relationship between observed NH_4-N and simulated wet NH_4-N deposition per
233 period is strong: GEOS-Chem performed well in simulating NH_4-N deposition across urban
234 areas (R^2 = 0.79; Figure 2). Although the regressions with (R^2 = 0.80) and without (R^2 = 0.79)
235 dry deposition of particulate NH_4-N had a similar R^2 value, inclusion of particulate NH_4-N

resulted in a slight improvement in normalized mean bias (NMB decreased from -44 to -41%). For most sites, the model underestimated observed NH_4-N deposition, including the network sites with weekly (i.e., MeMe) and monthly (i.e., ArBu, ArPe, UrMo, UrPa) sampling frequency. At the site level, the model did not capture interannual variation in NH_4-N deposition rates.

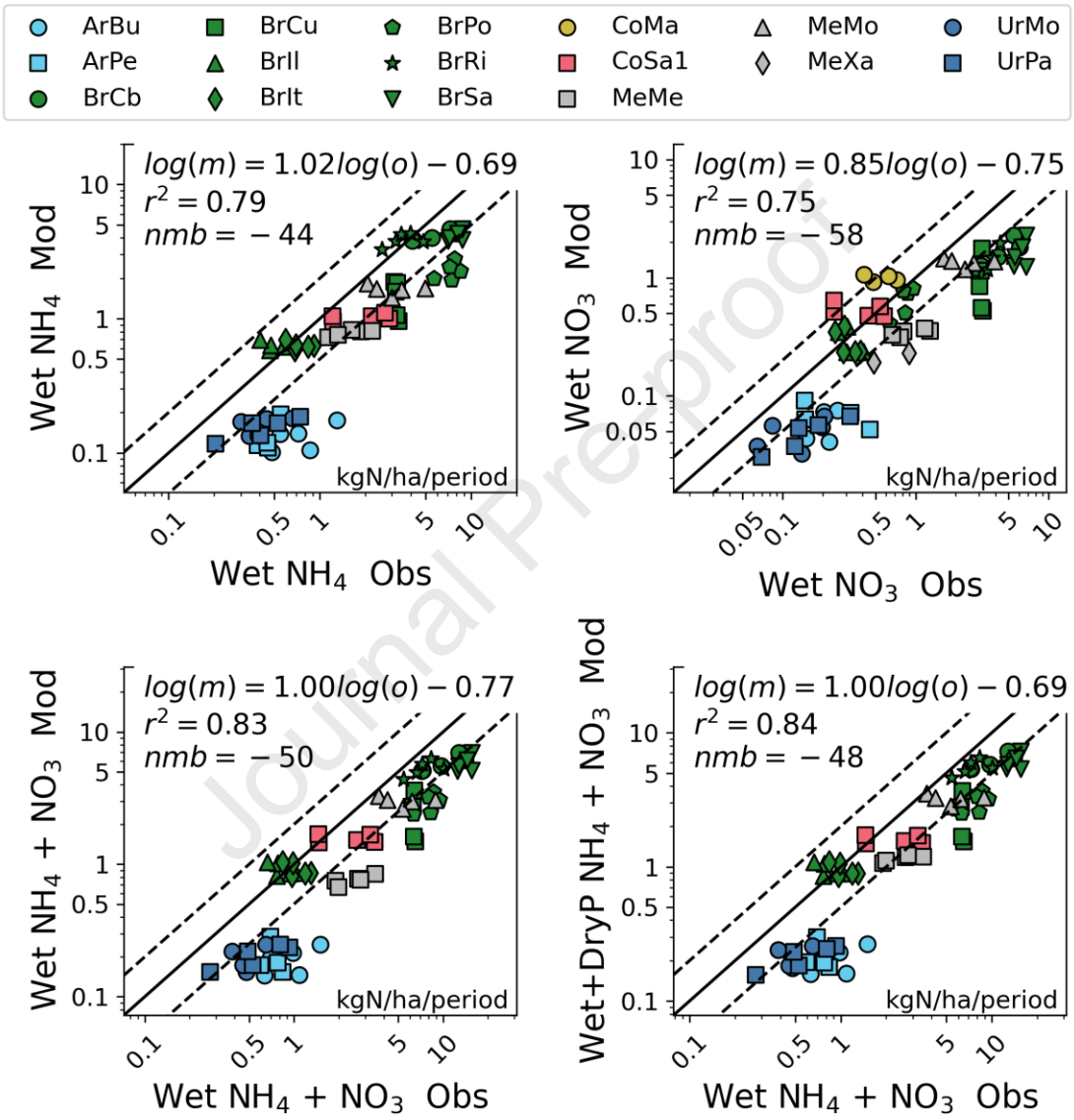

Figure 2. GEOS-Chem simulated wet N deposition versus observed N deposition to urban areas in Latin America. Individual symbols show N deposition per period (monthly to annual) for each year in the study window (2006-2010). Relationships are displayed for NH_4-N (top left; *n*=14 sites), NO_3-N (top right; *n*=16 sites), and inorganic N (NH_4-N + NO_3-N) deposition (bottom left; *n*=14 sites), and for GEOS-Chem simulated wet plus dry particulate inorganic N deposition

246 versus wet inorganic N deposition (bottom right; *n*=14 sites). Ammonium-N was not measured at
247 Manizales, Colombia (CoMa) or Xalapa, Mexico (MeXa). R^2 indicates the strength of the
248 relationship across urban areas. Dashed lines show the 2:1, 1:1, 1:2 performance.
249
250 GEOS-Chem performed less well for NO_3-N than for NH_4-N, yielding a lower R^2 value
251 of 0.75 and a more negative normalized mean bias of -58% (Figure 2). The model
252 underestimated NO_3-N deposition at most of the sites. Inclusion of dry particulate NO_3-N in the
253 model had no effect on the strength of the relationship between observed and simulated NO_3-N
254 or the normalized mean bias.
255 Overall, the relationship between observed and simulated N deposition was best for wet
256 inorganic N (Figure 2). The regression between observed inorganic N and wet plus dry
257 particulate inorganic N was nearly identical to that between observed and simulated NH_4-N
258 given the dominance of NH_4-N in atmospheric wet N deposition.
259 GEOS-Chem exhibited mixed performance for precipitation (Figure 3). However, the
260 model performed better for precipitation than for N across sites. The relationship between
261 observed and simulated precipitation had a higher R^2 (0.87) and insignificant bias (NMB = -1%).
262 Similar to N, the model did not capture interannual variation in precipitation.
263

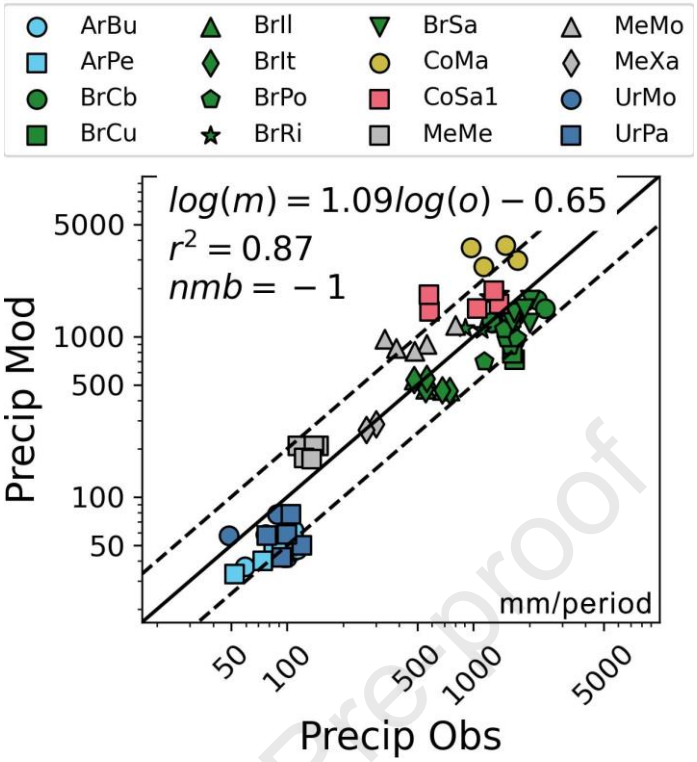

Figure 3. GEOS-Chem simulated precipitation (mm) versus observed precipitation in urban areas in Latin America. Individual symbols show precipitation per period (monthly to annual) for each year in the study window (2006-2010). R^2 indicates the strength of the relationship across urban areas. Dashed lines show the 2:1, 1:1, 1:2 performance.

3.2 Spatial patterns of inorganic N deposition

Observed mean annual inorganic N deposition for sites with a minimum of one year of observations ranged from 5.7-14.2 kg N ha^{-1} yr^{-1}, with a mean of 8.7 ± 0.8 kg N ha^{-1} yr^{-1} across all sites. In general, NH_4-N was the dominant form of N deposited (3.2-9.4 kg ha^{-1} yr^{-1}), comprising 48-90% of observed inorganic N (Figure 4). Nitrate-N deposition ranged from 0.8-6.2 kg N ha^{-1} yr^{-1}. Spatial patterns of deposition were reproduced better for NH_4-N, and for inorganic N given its dominance in atmospheric deposition, than for NO_3-N (Figure 4). Consistency between observed and simulated N deposition varied by N form and by region. For example, observed NH_4-N deposition was similar to simulated estimates of wet plus dry particulate NH_4-N deposition along the coast of Brazil. Urban areas, such as Rio de Janeiro and São Paulo, Brazil, with high observed NH_4-N deposition fell within grid cells with high simulated NH_4-N. This was not the case for NO_3-N, where there were larger discrepancies

between observed and simulated deposition. At the lower end of the range, GEOS-Chem performed well for the coastal Brazilian cities of Ilhéus and Itabuna, and Monterrey, Mexico. Interestingly, inconsistencies between observed and simulated estimates were largest for the network sites in Argentina and Uruguay with monthly sampling frequency and four to five years of continuous data. While both the model and the observations showed wet NH_4/NO_3 deposition ratios >1 in urban areas, the model often (n=8) overestimated the amount of NH_4 relative to NO_3. In only three urban areas did the model underestimate the NH_4/NO_3 ratio in wet deposition (Buenos Aires, Argentina, Porto Alegre, Brazil, and San Jose, Costa Rica).

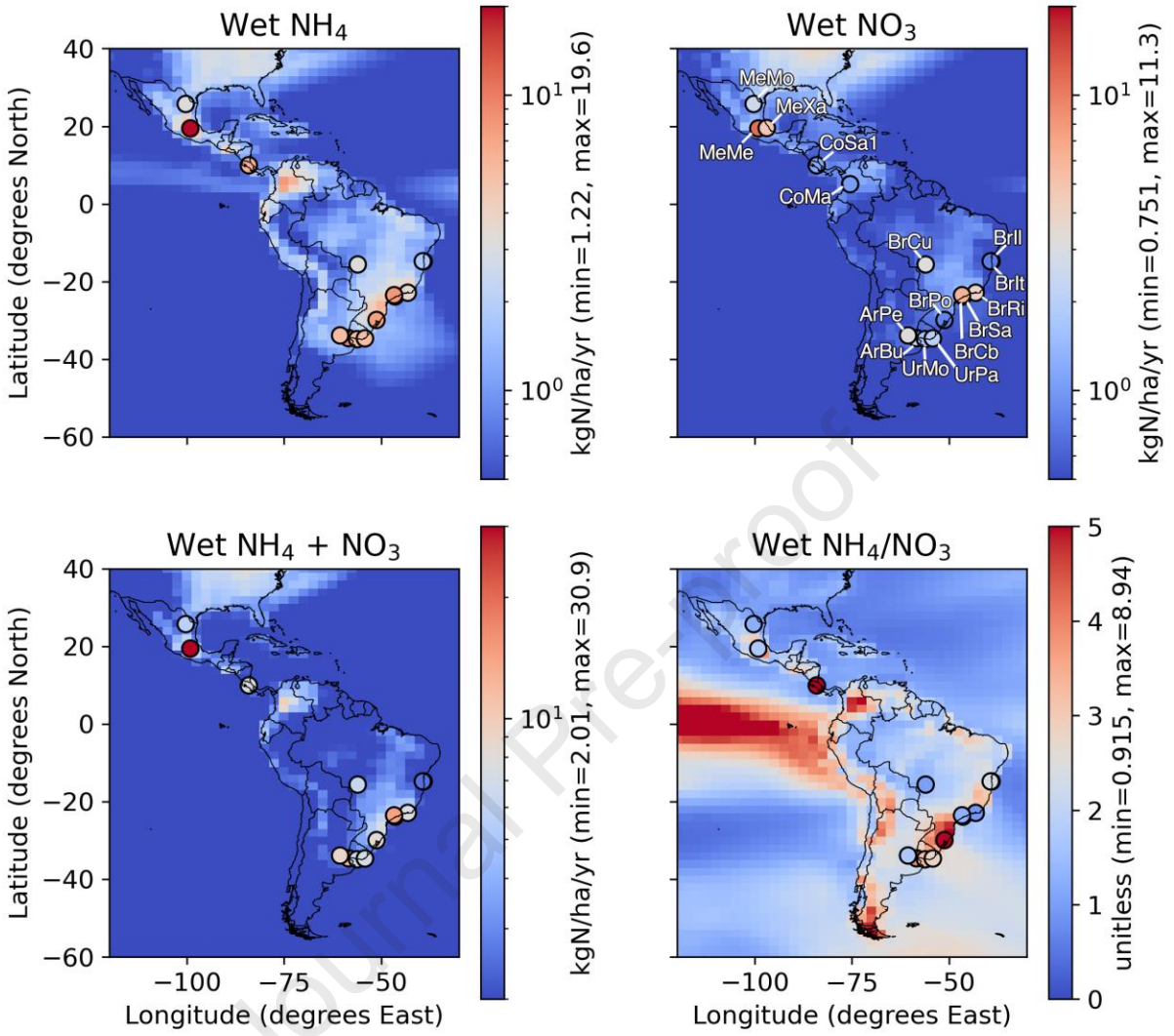

Figure 4. GEOS-Chem model output for mean annual wet plus dry particulate NH_4-N deposition (top left); wet plus dry particulate NO_3-N deposition (top right); wet plus dry particulate inorganic N (NO_3-N + NH_4-N) deposition (bottom left); and the ratio of NH_4-N/NO_3-N in wet plus dry particulate deposition (bottom right). Dots show observed mean annual N deposition to urban areas in Latin America for the period 2006-2010. Y-axis values include deposition rates that were scaled to enable plotting (see text for details). Ammonium-N was not measured at Manizales, Colombia (CoMa) or Xalapa, Mexico (MeXa). Dots for São Paulo and Cubatão and for Itabuna and Ilhéus overlap.

4. Discussion

Nitrogen deposition in urban Latin America is expected to increase in the future (Galloway et al. 2021). Yet, rates of N deposition are not yet known for many urban areas, and ecosystem responses to N deposition are poorly understood (Ponette-González et al. 2014; Ometto et al. 2020). Our observational dataset shows that mean annual wet N deposition to a cross-section of 16 urban areas is 8.7 ± 0.8 kg N ha^{-1} yr^{-1} and that GEOS-Chem performed reasonably well in capturing spatial variability in wet N deposition across these sites. Moreover, simulated mean annual dry N deposition is ~30-40% of total (wet plus dry) inorganic N deposition, indicating that ecosystems in urban areas likely receive a total N deposition load nearly double that of wet N deposition. Taken together, our results suggest that for the Latin American region, GEOS-Chem can be used to fill in important spatial and temporal gaps resulting from lack of N deposition measurements, provide dry deposition values and a more complete characterization of the N deposition budget, and provide source apportionment information that may be useful for control strategies.

Although GEOS-Chem v8-03-02 reproduced spatial patterns of N relatively well, the model generally underestimated N deposition to urban areas compared to observations. Additional measurements of N deposition are needed to elucidate whether this pattern is consistent across Latin American urban areas, which encompass more diverse geographic and climatic contexts and ecological surroundings than the sites in our dataset. What could be driving GEOS-Chem underestimates of wet N deposition? Several factors, including meteorology, characteristics of the observational dataset, emissions estimates, and process representation in this model version may have contributed to model bias. For meteorology, the most obvious effect would be a bias in precipitation, however our comparisons revealed that the model performed best for precipitation. From this, we infer that the relationship between observed and simulated N deposition was more strongly influenced by other factors. For instance, even though gaseous and particulate N deposition to bulk collectors is thought to be minimal (Cook et al. 2018), it is possible that bulk deposition collectors, which were deployed at most of the sites, may have collected some amount of dry deposition (Fenn et al. 2009). At the RP-RainNet sites, comparisons show higher NO_3^- inputs to bulk than wet-only collectors (Michel et al. 2020). We considered the potential for dry-deposition artifacts (after Dämmgen et al. 2005) by including particulate N (~5% of total dry N deposition) in the model simulations. While there was a slight

improvement in the normalized mean bias, we suspect that the lack of high temporal resolution data on volume-weighted N concentrations was more important.

A scale mismatch between the grid average land cover represented by the model and the land cover at the sampling location likely also contributed to underestimation of N deposition by GEOS-Chem. Nitrogen in rainwater was frequently sampled at one to few highly urbanized locations whereas N deposition was simulated using coarse grid cells with varying fractions of urban land cover. Presumably, the latter would artificially dilute NO_x and NH_3 emissions from vehicles and thus underestimate wet N deposition.

We were surprised to find that the model performed least well for urban areas in Argentina and Uruguay that are part of atmospheric deposition monitoring networks with monthly collection of rainwater samples. Livestock is a major source of NH_3 emissions in this region. Although the model included livestock emissions, uncertainties in Latin American NH_3 emissions inventories are the most likely explanation for the gross underestimate of NH_4-N deposition. Also, the rapid increase in emissions from both the agricultural and livestock sectors (Castesana et al. 2018), not accurately captured in global emission inventories used as input to our model calculations, would further contribute to model underestimation.

Finally, we evaluated the potential influence of model version and emissions inventories on our results by comparing wet N deposition simulated using GEOS-Chem version 08-03-02 and version 11-01 (Ackerman et al. 2019). Ackerman et al. (2019) used a newer version of the model and EDGAR v4.2 for NH_3 and NO_x. The only year simulated by both studies is 2006, which we compared. Figure S1 shows the spatial distribution of differences, and the text describes percent differences at the sampling locations. Rates of total N deposition were comparable between versions with a median difference of +9% and ranging from -23% to 43%. Note, however, that the wet deposition is most important for comparison to observations. The wet deposition in Ackerman et al. was higher with a median difference of +28% at our sampling locations and ranging from -52% in Mexico City to 119% in Uruguay. In contrast, their dry deposition was lower than this work with a median difference of -13% and ranging from -47% to +129%. Because wet deposition dominates the measurements (dry particles adjustment contributed very little), this suggests that using the newer version of the model and the emission inputs would improve underestimates of the observations.

5. Conclusions

We conclude that despite the limitations noted above, GEOS-Chem provides a reasonable representation of N deposition across Latin America in the absence of dense observations, and for urban areas where atmospheric N pollution is high and increasing. Moreover, our analysis suggests that the model can be used to fill in important gaps in ecosystem N budgets in a region undersampled for N inputs. Additional observations of N deposition in Latin America, through the Nitrogen Human Environment Network (Nnet) for example (Ometto et al. 2020), will provide opportunities to further constrain model estimates of N deposition in the future.

Disclaimer

The views expressed in this article are those of the authors and do not necessarily represent the views or policies of the U.S. Environmental Protection Agency.

Author Contributions

Ponette-González: Conceptualization, Formal analysis, Writing – Original; **Lewis**: Methodology, Investigation, Writing – Review & Editing; **Henderson**: Conceptualization, Methodology, Software, Formal analysis, Resources, Writing – Review & Editing, Visualization; **Carnelos**: Investigation, Resources, Writing – Review & Editing; **Piñeiro**: Investigation, Resources, Writing – Review & Editing; **Weathers**: Conceptualization, Writing – Review & Editing; **Schwede**: Conceptualization, Methodology, Writing – Review & Editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data Availability

The dataset related to this article can be found in the Supplementary Information and will be hosted on the Knowledge Network for Biocomplexity.

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Highlights

- GEOS-Chem simulated wet inorganic N deposition is evaluated for urban Latin America.
- Across urban areas, GEOS-Chem captures spatial variability in N deposition well.
- At the site level, the model does not capture year-to-year variation.
- Observed inorganic N deposition to 16 urban areas ranged 5.7-14.2 kg ha^{-1} yr^{-1}.

Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: