Nitrous Oxide Emissions from Municipal Wastewater Treatment

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Nitrous Oxide Emissions from Municipal Wastewater Treatment

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Nitrous oxide (N₂O) emissions from primary and
secondary wastewater treatment processes were
measured during spring and summer 1993 in Durham,
NH. The most significant emissions occurred during
secondary aeration. Dissolved N₂O generated as a
result of denitrification during primary settling was
stripped from the liquid during mechanical aeration.
Emission factors derived from our field measure-
ments included per capita emissions of 3.2 g of N₂O
person^{-1} yr^{-1} and flow based emissions of 1.6 ×
10^{-6} of N₂O (L of wastewater)$^{-1}$.

Introduction

Nitrous oxide (N₂O) is an important trace gas that has been
identified as a significant constituent in several aspects of
atmospheric chemistry. It is important as a greenhouse
gas due to its efficiency in absorbing infrared radiation (1).
N₂O is also a major source of nitrogen oxides to the
stratosphere where it plays a catalytic role in ozone
destruction (2). The current atmospheric mixing ratio of
N₂O is about 310 ppbv and is increasing at a rate of 0.25–
0.31% yr^{-1} (3).

A key to formulating strategies to control and reduce
the rate of increase of N₂O in the atmosphere is the
identification and quantification of all sources, both natural
and anthropogenic. Significant natural sources include
undisturbed soils, oceanic waters, and possibly atmospheric
formation. These sources account for about 60% of total
global N₂O emissions, which are estimaed to range from
17 to 30 Tg of N₂O yr^{-1} (3, 4). Many anthropogenic sources
have also been identified but remain poorly quantified.
These sources include cultivated soils, biomass burning,
combustion, industrial processes, and waste treatment (4).
The current rate of increase of the global N₂O mixing ratio
implies a 40% excess of sources over sinks (5). Anthro-
pogenic sources present an opportunity to reduce this
excess, but better quantification is necessary to establish
a source hierarchy to guide the application of mitigation
resources.

The purpose of this study was to quantify in-system
N₂O emissions from a municipal wastewater treatment
plant consisting of processes typical of those in use in the
industrialized world. The transport and management of
residential, commercial, and industrial wastewater is known
to produce N₂O (3). The dominant mechanism of N₂O
production in these systems has recently been identified

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as bacterially mediated denitrification (6, 7). Denitrification
occurs under anaerobic conditions, which are characterized
by low oxygen partial pressure. These conditions typically
occur in wastewater when aerobic decomposition processes
demand more oxygen than can be supplied by surface
diffusion. Therefore in the absence of mechanical aeration,
denitrifying bacteria activate the anaerobic conversion of
NO₃$^-$ to N₂ gas and produces N₂O gas as an intermediate
byproduct.

McElroy et al. described the enhancement of N₂O
emissions from the surface waters of a tidal river system
affected by the release of primary-treated wastewater and
sludge (8). However, few quantitative data exist regarding
the generation of N₂O in processes that comprise modern
wastewater treatment plants. This lack of data will limit
further technical assessments of mitigation options. The
field measurements described below were performed to
quantify N₂O emissions from processes in a wastewater
treatment plant and to compare the field-derived N₂O
emission factor to emission factors derived from recent
laboratory investigations by Debruyn et al. (6).

Field Site

The municipal wastewater treatment plant studied is located
in Durham, NH. The facility serves the town and the main
campus of the University of New Hampshire. The popula-
tion served during the school year is approximately 12 500
and generates a mean wastewater flow of approximately 4
× 10⁶ L day^{-1}. The population served during the summer
is approximately 6200 and generates a mean wastewater
flow of approximately 2 × 10⁶ L day^{-1}. During the summer
months, the plant processing capacity is normally reduced
by one-half through partial shut down of the grit tanks,
primary settling tanks, secondary aeration tanks, and
secondary settling tanks.

The facility consists of primary and secondary unit
operations and processes (Figure 1) that treat a medium-
strength municipal wastewater with an average influent
BOD₅ at 20 °C of approximately 250 mg L^{-1} with an
estimated suspended solids concentration of 220 mg L^{-1}.
Average removal efficiency is 94%. A set of two grit
chambers is the first physical unit operation encounterd
by the influent where sand, gravel, and other heavy solids
are removed from the system. The wastewater then enters
the primary settling tanks where it is held in a relatively
quiescent state resulting in the removal of solids with a
higher specific gravity than the liquid by settlement and
solids with a lower specific gravity by flotation and
skimming. The liquid then enters secondary treatment,
while the solids are removed from the system as primary
sludge. The secondary treatment system consists of four
aeration tanks and two clarification tanks. In this process,
the wastewater organic matter that enters the aeration tanks
is decomposed by suspended bacterial biomass under
aerobic conditions. This results in the release of gases to
the atmosphere and the growth of microbial biomass. The
microbial-rich liquid then passes into the secondary
clarification tanks where the microbial biomass is separated
from the treated wastewater by gravity. A portion of the
settled microbes is continuously recycled back to aeration
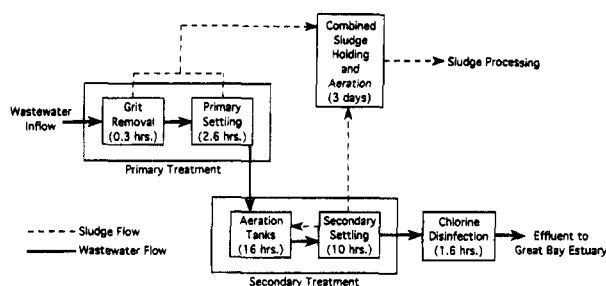tank 1 as an inoculum to maintain a proper concentration

FIGURE 1. Simplified process diagram for the Durham, NH, wastewater treatment plant. Retention times are given in parentheses.

of microorganisms in the aeration tanks. The remaining fraction of the microbial biomass is treated as sludge, which is periodically diverted to a holding tank depending on load conditions. The mixed sludge is then transferred to the sludge processing building where it is dewatered and prepared for disposal. The treated effluent moves to the chlorine contact tank for disinfection and is released into the Great Bay Estuary.

Methods

Nitrous oxide fluxes were measured from each tank in the wastewater treatment plant during a preliminary evaluation of plant emissions. The dominant emission sources were determined to be the grit tanks, secondary aeration tanks, and sludge holding tanks. Negligible emissions were measured from the surface of the primary settling tanks and the secondary clarification tanks. N_2O fluxes were then measured from the dominant sources at roughly 1-week intervals at approximately the same time of day. Gas flux measurements were collected from aerated and nonaerated liquid surfaces. The spatial variability of measured N_2O concentration was examined early in the study in each source tank. The maximum measured spatial variation was found in the aerated area of the grit tank. This variability was determined to be of negligible impact when compared to the expected temporal variability of emissions. The sample collection procedures employed are described below.

Nonaerated Surfaces. A closed chamber technique was used to measure fluxes from nonaerated liquid surfaces (9). An aluminum enclosure (51 × 51 × 26 cm) was floated on the liquid surface with a thermistor mounted inside the enclosure to measure the enclosed air temperature. Floats were fastened to the enclosure sides, and the chamber was manually held in place during sampling to minimize chamber movement due to surface turbulence. A battery-operated blower circulated the air within the enclosure and through a closed loop of hose from which headspace air samples were withdrawn. Samples were collected in 20-mL nylon syringes at 2-min intervals for 10 min. The gas flux, E (Mass area^{-1} time^{-1}), from the liquid surface in the static chamber was calculated by

$$E = (V/A)p(dc/dt) \qquad (1)$$

where V is the volume of the chamber, A is the enclosed surface area, p is the density of the gas at the temperature recorded in the chamber, and dc/dt is the linear increase in the gas concentration in the chamber during the sampling period.

Aerated Surfaces. A bag technique was used to measure fluxes from aerated liquid surfaces. A 40-L polyethylene

sample bag, fastened to the inside of a wood support frame, was used to collect gas samples from the agitated surface of the aerated tanks. The open base of the frame allowed gas to be collected from 0.084 m^2 of the surface. To collect samples, the bag was collapsed and emptied of ambient air, and the wood support frame was placed several inches into the liquid. Dissolved gases within the liquid, transferred to the mechanically distributed air by diffusion, then filled the collection bag. Samples were withdrawn from the bag in 20-mL nylon syringes. The gas flux, E (mass area^{-1} time^{-1}), was calculated by

$$E = pcQ/A \qquad (2)$$

where p is the density of the gas at the temperature recorded in the sample bag, c is the sample gas concentration, Q is the total diffuse air flow, and A is the total surface area.

Sludge Gases. N_2O fluxes resulting from sludge storage were determined by two methods. First, gas concentrations in the sludge were determined by sampling the sludge flow, prior to transfer, with a 0.5-L collection bucket. Samples of 30 mL of sludge were collected immediately after withdrawal from the flow in 60-mL polypropylene syringes. An equal volume of laboratory room air was later introduced into the sludge sample syringe, and the syringe was shaken vigorously for 2 min. The gases in the syringe headspace were then transferred to a clean 20-mL nylon syringe for analysis. This procedure was repeated five times and stripped greater than 95% of the dissolved N_2O from the sludge samples. This method estimated the quantities of gases emitted during, and immediately after, transfer of sludge to the holding tanks. Fluxes were calculated under the assumption that the total gas potential of the sludge was realized by aeration stripping during the period of storage prior to final processing.

Second, fluxes resulting from the generation of gases within the sludge holding tanks, and stripped from the liquid during aeration, were determined using the aerated surface collection method described previously. This flux was assumed to be continuously present as a secondary background flux. Emissions from the surface of the nonaerated area of the sludge storage tanks were also evaluated using the closed chamber technique described previously.

Gas Analysis. All samples were analyzed as described by Keller et al. (10). Samples were immediately returned to the lab, allowed to equilibrate to laboratory temperature for 1–2 h, and analyzed for N_2O within 3–4 h after collection. Samples and standards were dried across $CaSO_4$ as they were injected into the gas chromatograph (GC) injection loop. Analyses for N_2O were accomplished using an electron capture detector (ECD-GC) after constituent separation using 2 m × 3.2 mm o.d. columns packed with Hayesep N. Carrier gas was a 95% Ar/5% CH_4 mixture. Peaks were quantified with Hewlett Packard Chemstation software on a personal computer. N_2O standard gases were obtained from Scott Specialty Gases and were calibrated with a National Institute of Standards and Technology certified standard. Precision of analysis for calibration gases was 0.3%.

Statistical Analysis. Emissions data were examined for statistical distribution to determine the appropriate form of statistical analysis. N_2O flux data from the secondary aeration tank 1 was found to be log-normally distributed, requiring the application of parametric statistics to the data
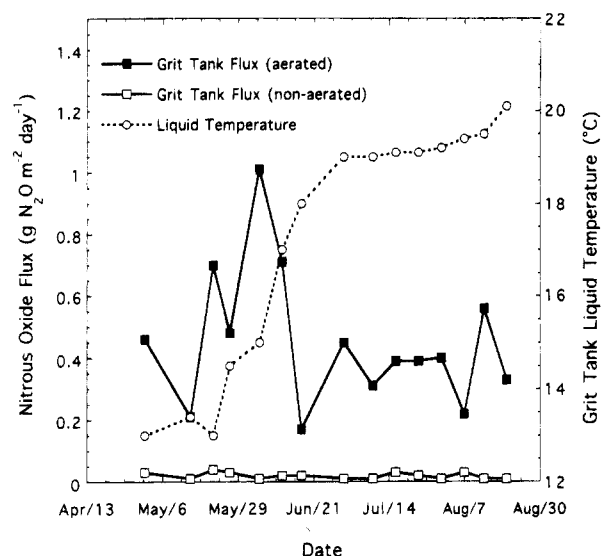
FIGURE 2. Observed N₂O fluxes (g of N₂O m⁻² day⁻¹) and wastewater temperatures (°C) from the aerated and nonaerated areas of the grit tank.

in log-transformed form. All other sample data were determined to have been drawn from normally distributed populations, permitting application of parametric statistics. Relationships between emissions and temperature were examined using linear regression techniques (11).

Results

Grit Tanks. N₂O fluxes from the aerated area of the grit tanks, measured under normal operating conditions, are presented in Figure 2. Fluxes ranged from a minimum of 0.17 g of N₂O m⁻² day⁻¹ to a maximum of 1.05 g of N₂O m⁻² day⁻¹. A maximum flux of 2.48 g of N₂O m⁻² day⁻¹ was measured in the aerated area of the grit tanks after the disposal of a septic tank load by a local septage hauler. These septage disposal events occur several times during a typical week, at which time gas emissions are elevated. However, these events last only a single detention period and do not contribute significantly to total weekly emissions and were disregarded.

N₂O fluxes from the nonaerated area of the grit tanks, measured under normal operating conditions, are also presented in Figure 2. Fluxes ranged from a minimum of 0.01 g of N₂O m⁻² day⁻¹ to a maximum of 0.04 g of N₂O m⁻² day⁻¹. Higher fluxes were also measured in this area during the septage disposal events described previously but are not representative of normal plant operations and were disregarded.

The N₂O emissions data from the grit tanks were examined for temperature dependence by linear regression analyses. No statistically significant correlation was observed in either the aerated areas (r^2 = 0.13) or the nonaerated areas (r^2 = 0.16).

Secondary Treatment Tanks (Aerated). Wastewater from the primary settling tanks and return sludge enter the secondary aeration system in tank 1. Preliminary measurements established tank 1 as the significant N₂O source in the four-tank aeration system. Samples collected from tanks 2—4 were consistently less than 10% of fluxes measured in tank 1 and were therefore neglected in total plant emission calculations. Gases generated and dissolved in the liquid during primary settling, as well as gases generated in the return sludge during settlement in the
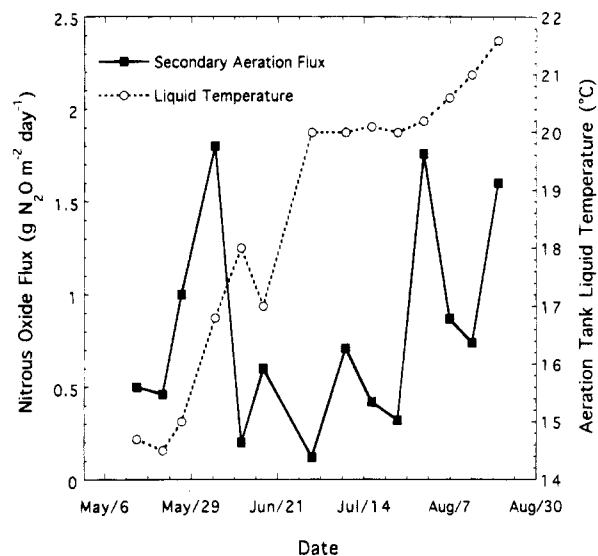

FIGURE 3. Observed N₂O fluxes (g of N₂O m⁻² day⁻¹) and wastewater temperatures (°C) from secondary aeration tank 1.

secondary clarification tanks, are later stripped from the liquid column by aeration. N₂O fluxes from tank 1 ranged from a minimum of 0.12 g of N₂O m⁻² day⁻¹ to a maximum of 1.80 g of N₂O m⁻² day⁻¹ (Figure 3).

The aeration tank N₂O emissions data were examined for temperature dependence by linear regression analyses. No statistically significant correlation was observed in either of the aerated areas (r^2 = 0.11).

Sludge Holding Tanks. Approximately 2.4 × 10⁶ L of sludge from the primary and secondary treatment processes are transferred to the sludge holding tanks annually. The sludge is aerated in the holding tank for several days to reduce the liquid content prior to transfer to the sludge processing facility. Gas samples collected from the aerated surface early in the study indicated highly variable fluxes dependent on the time since sludge transfer. Fluxes from the aerated surface as high as 3.1 g of N₂O m⁻² day⁻¹ were detected immediately after transfer to the holding tanks, while a flux of 0.24 g of N₂O m⁻² day⁻¹ was measured from a batch of sludge after 1 day of aeration.

The flux potential of gases dissolved in the sludge was estimated by determining total dissolved N₂O per unit volume of sludge and was assumed to be fully realized. N₂O concentrations ranged from a minimum of 15.3 μg of N₂O (L of sludge)⁻¹ to a maximum of 145 μg of N₂O (liter of sludge)⁻¹.

Gas fluxes from stored sludge were measured from the aerated areas of the storage tanks to determine the flux of gases due to production during storage. Only samples collected a minimum of 1 day after sludge transfer were considered appropriate to estimate this secondary flux. A minimum of 1 day of aeration was considered necessary for the depletion of gases dissolved in the sludge during accumulation and transfer. Data collected on six sampling dates met this criteria. N₂O fluxes ranged from a minimum of 0.13 g of N₂O m⁻² day⁻¹ to a maximum of 0.28 g of N₂O m⁻² day⁻¹. The nonaerated areas of the storage tanks were also examined as a possible emissions source. Fluxes from the nonaerated areas were found to be less than 10% of those from the aerated areas and were disregarded.

Discussion

Nitrous oxide is generated by biologically mediated denitrification in wastewater treatment systems in areas of high

biological oxygen demand and low oxygen concentration such as the influent lines, primary settling tanks, secondary clarification tanks, sludge holding tanks, and sludge transfer lines. The full potential of dissolved N_2O is realized in areas of mechanical aeration where gases are stripped from the liquid by diffusion. In the Durham plant, N_2O generated in the influent lines was emitted in the aerated areas of the grit tanks by aeration and in the nonaerated areas as the result of turbulent liquid flow initiated by aeration.

N_2O generated in the primary settling tanks and in the returned sludge was stripped by aeration from the liquid in the secondary aeration tanks. The highest emissions of N_2O from the secondary aeration system were measured in the first tank where the return sludge and wastewater enter the system. N_2O emissions decreased significantly as the liquid moved sequentially through the four-tank process and were negligible after transfer of the liquid to the secondary clarifiers.

N_2O generated in the wasted sludge during storage was emitted during aeration in the holding tanks immediately after transfer. Additional N_2O was generated in the sludge in the nonaerated areas of these tanks during the 3-day detention period and was stripped from the liquid column as it flowed into the aerated areas.

A significant correlation between methane (CH_4) emissions and temperature was observed in the wastewater treatment processes at this facility as previously reported (12). The absence of a temperature correlation with N_2O emissions is therefore surprising due to the assumed microbial production of both CH_4 and N_2O in these environments. Greater than weekly sampling frequency, sampling during winter low temperatures, and oxygen gradient measurements may be necessary to accurately characterize the relationship between temperature and N_2O emissions in this system.

The mean flux of N_2O measured from the aerated area of the grit tanks was 0.45 g of N_2O m^{-2} day^{-1}. During the local university school year, from September through May, two grit tanks are operated in parallel with a total aerated surface area of 11.2 m^2. During the summer months, from June through August, wastewater flow and organic load are reduced by approximately half due to the local population reduction. Primary treatment capacity during the summer low flow period is subsequently reduced by the shutdown of one grit tank and the total aerated surface area drops to 5.6 m^2. These reductions preserve the continuity of emissions from the grit tank, requiring only an area weighting in total annual flux calculations. The population weighted area is 9.8 m^2. The estimated total area weighted annual emissions from the aerated area of the grit tanks were 1.6×10^3 g of N_2O yr^{-1}.

The mean flux measured from the nonaerated area of the grit tanks was 0.02 g of N_2O m^{-2} day^{-1}. Total nonaerated surface area was 26 m^2 from September through May and 13 m^2 from June through August, yielding a population weighted surface area of 22.8 m^2. The estimated total area weighted annual emission from the nonaerated area of the grit tanks was 158 g of N_2O yr^{-1}.

The mean N_2O flux from secondary aeration tank 1 was 0.61 g of N_2O m^{-2} day^{-1} while emissions from the remaining tanks were negligible. All four secondary aeration tanks remained operational throughout the sample collection period and are normally operational throughout the year with single-tank aerated surface areas of approximately 144

TABLE 1

Process and Total N_2O Fluxes (of N_2O yr^{-1}) from the Durham, NH, Wastewater Plant

process	N_2O flux
grit tank (aerated)	1.6×10^3
grit tank (nonaerated)	1.6×10^2
aeration tanks	3.2×10^4
sludge storage tank	1.7×10^3
total	3.5×10^4

m^2. The estimated total annual emissions from secondary aeration were 3.2×10^4 of N_2O yr^{-1}.

The mean sludge sample gas concentration was 105 μg of N_2O (L of sludge)$^{-1}$. Approximately 2.4×10^6 L of sludge is transferred annually to the sludge holding tanks for aeration. Assuming complete removal of dissolved gases by aeration yields annual fluxes from sludge transfer of 252 g of N_2O yr^{-1}.

The mean secondary N_2O flux from the aerated areas of the sludge holding tanks was 0.21 g of N_2O m^{-2} day^{-1}. Two of the three holding tanks were typically utilized for sludge storage on a continuous basis. The total aerated surface area was 18.6 m^2, yielding annual fluxes from sludge storage due to secondary production of 1.4×10^3 g of N_2O yr^{-1}. Total estimated annual flux from sludge transfer and storage was 1.7×10^3 g of N_2O yr^{-1}.

The estimated total process N_2O flux is summarized in Table 1. The total estimated annual N_2O flux from the Durham, NH, wastewater treatment plant is 3.5×10^4 g of N_2O yr^{-1}.

A weighting must be applied to calculate the population served by the wastewater treatment plant due to annual variations in the number of town residents. Approximately 12 500 people from September through May and approximately 6200 from June through August generate wastes that are treated at this facility. The time-weighted average of this population is 10 925 people, which yields a per capita gas flux of 3.2 g of N_2O person^{-1} yr^{-1} from wastewater treatment plants with typical primary and activated sludge treatment processes.

Flow-based emission rates can also be determined. Based on an annual wastewater flow of 1.1×10^9 L yr^{-1}, annual gas flux is estimated to be 1.6×10^{-6} g of N_2O (L of wastewater)$^{-1}$ for the primary treatment system and 3.1×10^{-5} g of N_2O (L of wastewater)$^{-1}$ for the activated sludge secondary treatment system.

Total annual N_2O emissions from wastewater treatment in the United States can be estimated using our emission factors and national wastewater and sludge handling statistics. Wastewater flow through primary treatment in the United States in 1988 was estimated to be 4.1×10^{13} L yr^{-1} (13). This study also found return activated sludge to be the most widely used method of secondary treatment with a flow of 3.9×10^{13} L year^{-1}. Total N_2O emissions from these systems, based on the Durham wastewater treatment plant emission factors, are estimated to be 6.6×10^7 g of N_2O yr^{-1} from primary treatment and 1.2×10^9 g of N_2O yr^{-1} from secondary activated sludge treatment.

A recent laboratory investigation of N_2O generation potential from wastewater has resulted in emission factors for raw wastewater and wastewater after primary sedimentation. Wastewater samples were incubated in the laboratory yielding weighted average temperature emission

factors of 23 ± 21 μg of N_2O (g of ss)$^{-1}$ for raw wastewater and 770 ± 170 μg of N_2O (g of ss)$^{-1}$ for effluent treated by primary sedimentation, where g of ss stands for "gram of suspended solids" (6). These values can be compared to our results by converting our volume-based emission factors to suspended solids-based emission factors. Assuming a suspended solids concentration for untreated medium-strength domestic wastewater of 220 mg L^{-1} (14) yields emission factors of 7.3 μg of N_2O (g of ss)$^{-1}$ for raw wastewater and 132 μg of N_2O (g of ss)$^{-1}$ for effluent entering the secondary aeration system. These values compared well with the results of Debruyn et al. Higher observed emission factors from the laboratory incubations of Debruyn et al. may have resulted from the inclusion of higher strength industrial wastewater samples and ideal incubation conditions.

Our results are the first step to a quantitative national inventory of N_2O emissions from wastewater treatment and should be viewed as an approximation of the order of magnitude of these emissions. Sampling of a wide variety of wastewater streams, treatment technologies, and operating conditions will be necessary to better refine these estimates. The treatment of industrial wastewater appears to hold the greatest potential for uncertainty in emission estimates. Global industrial sources generated approximately 4.74×10^{14} L of wastewater in 1990 with organic matter contents, measured as BOD$_5$, ranging from 1000–35000 mg L^{-1} (15). Organic matter of this volume and concentration represents a significant potential for denitrification and N_2O generation depending on the treatment methods utilized. For example, wastewater and sludge lagoons often operate under facultative or anaerobic conditions, creating an ideal environment for denitrification. These systems are used worldwide for the treatment of industrial as well as domestic wastewater. Determining how much of this N_2O emission potential is actually realized will require continued field measurements and better characterization of global wastewater management practices.

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