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Invited Paper

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3	On the use of data from commercial NOx analyzers for air pollution studies
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9	
10	Abstract
11	Reactive oxidized nitrogen species play a central role in environmental pollution, and long term
12	monitoring is widespread. But conventional NOx (defined as NO + NO ₂) analyzers employing
13	heated converters respond to many species in addition to the compounds NO and NO ₂ . The
14	response of these instruments to nitric acid, peroxyacetyl nitrate, alkyl nitrates and other oxidized
15	nitrogen species (the sum of these plus NOx is defined as NOy) is well established, but the ratio
16	of NOx to NOy varies widely in time and space making the accuracy of commercial NOx
17	monitors uncertain. Care must be taken when comparing spectroscopic measurements of NO2 or
18	numerical models to output from commercial NOx monitors. Correction factors can be
19	developed for specific conditions, and long term trends can be meaningful. Recent studies
20	comparing modeled NOx to measurements with large interferences can involve errors of a factor
21	of two or more and produce misleading guidance on science and policy; the need for rigorous
22	model evaluation adds urgency to the deployment of "true NOx" monitors.
23	
24	Introduction
25	Excess reactive oxidized nitrogen, concentrations above the natural atmospheric background in
26	the atmosphere, lead to a variety of environmental problems, especially ground level ozone, a
27	recalcitrant air quality problem that poses a threat to human health and climate. Ozone
28	production rates respond nonlinearly to NOx concentrations [<i>Chameides et al.</i> , 1992; <i>Crutzen</i> ,
29	19/3; Sillman et al., 1990] making it vitally important to measure NOx (NO+NO ₂) specifically
30	and to represent NOx concentrations accurately in chemical transport models e.g., [Lelieveld et
31	<i>al.</i> , 2015]. If the modeled concentration of NOX is wrong, simulations can give misleading
32	guidance on pollution control policy. Because ozone production rates fall off at higher NOx
33 24	herefit to ozono from a given out in NOx concentrations can contribute to underestimates in the
54 25	benefit to ozone from a given cut in NOX emissions e.g., [Guttana et al., 2008].
26	Oxides of nitrogen are most frequently monitored via a chemiluminescent reaction with ozone
30	[<i>Fontiin at al.</i> 1070] with NO measured directly and higher oxides following reduction to NO
30	[Folight et al., 1970] with NO incastical ancerty and higher oxides following reduction to NO $[F_{abs}anfald at al. 1087; Winar at al. 1074]. Because NO is designated one of the criteria$
30	pollutants by the United States Environmental Protection Agency (USEPA). States are required
10	to demonstrate attainment of the standard and chemiluminescence NOx analyzers are in
40 //1	common use Most monitors reporting to the EPA's Air Quality System AOS
41 42	(https://www.ena.gov/ags) employ commercial NOx analyzers with hot molybdenum NO ₂
42 43	$(nups.//www.epa.gov/aqs) employ connected it for analyzers with not mory denum NO_2converters – one purpose of this paper is to alert users these do not measure true NOx because$
44	they suffer substantial interferences from other reactive nitrogen species. These interfering
45	species include nitrous acid (HONO) nitric acid (HNO ₃) nitric acid anhydride (N ₂ O ₅) organic
46	nitrogen peroxides, alkyl nitrates (RONO ₂), nitryl chloride (CINO ₂) and other important air
	r = -6 - r

- 47 pollutants. These instruments more nearly measure NOy than NOx; in the US, a good
- 48 approximation is $NOy = NOx + HONO + HNO_3 + 2XN_2O_5 + PANs + RONO_2$, where PANs
- 49 represents the family of peroxyacetyl nitrates and RONO₂ represents the family of alkyl nitrates.
- 50 When nitrate aerosol passes through the inlet it can likewise be detected as NOx, and some
- 51 species such as NH_4NO_3 thermally decompose to NH_3 and HNO_3 that can cause interferences.
- Although NH₃ oxidation on heated molybdenum is usually small at ambient humidity, under
 certain circumstances ammonia and amines can also cause substantial interferences [Saylor et al.,
- 53 certain circumstances ammonia and ammes can also cause substantial interferences [*Saylor et al.* 54 2010; *Suzuki et al.*, 2011]. The difference between NOy and NOx is sometimes referred to as
- 55 NOz, and gives an indication of the aging of the air parcel e.g., [*Gaudel et al.*, 2018; *Kleinman et*
- 56 *al.*, 2002]. Commercial "NOx" instruments are in common use because the interferences do not
- 57 cause a problem when such monitors are deployed to demonstrate attainment with NO₂ standards
- they provide an upper bound on NO₂ and NOx concentrations. So while these monitors can be
- 59 useful, they generate numbers with variable, and often severe, high bias.
- 60
- 61 The high efficiency of hot molybdenum converters for NOy has been known for some time
- 62 [Fehsenfeld et al., 1987; McClenny et al., 2002] and many investigations have documented or
- 63 quantified substantial interferences in commercial instruments deployed in Europe, Asia, South
- 64 America, as well as North America [Dunlea et al., 2007; Geddes and Murphy, 2014; Hassler et
- 65 *al.*, 2016; *Lamsal et al.*, 2008; *Leston and Ollison*, 2017; *Luke et al.*, 1998; *Ordonez et al.*, 2006;
- 66 Piters et al., 2012; Poulida et al., 1994; Reed et al., 2016; Steinbacher et al., 2007; Suzuki et al.,
- 67 2011; *Villena et al.*, 2012; *Wild et al.*, 2014; *Xu et al.*, 2013]. In this invited paper we will
- 69 well as models and measurements of reactive nitrogen, and show how uncertain high bias or
- assumptions of equivalence between NOx and NOy can lead to misleading results.
- 71

72 State of the Science

- 73 A review of the literature demonstrates that the fraction of atmospheric NOy composed of NOx
- usually fall off quickly with distance from sources. Near emitters, NOx can dominate NOy in
 the winter in daytime [*Allen et al.*, 2018; *Salmon et al.*, 2018], but in the summer, when
- 76 photochemical smog production is at a maximum, NOx is usually a small fraction of the total
- 77 NOy even close to cities. For example, in DISCOVER-AQ using research grade instruments
- 78 over the Baltimore/Washington area, true NOx accounted on average for less than half the total
- 79 NOy [*Anderson et al.*, 2014; *Hembeck et al.*, 2019; *Lee et al.*, 2018]. Similar results were
- 80 observed in Michigan [*Thornberry et al.*, 2001], Texas (https://www-
- 81 air.larc.nasa.gov/missions/discover-aq/P3B-Profiles.tx2013.html), New York State [*Ninneman et*
- *al.*, 2019; *Schwab et al.*, 2009] and Switzerland [*Ordonez et al.*, 2006]. At roadside in heavy
- traffic most of the NOy can be NOx [*Hassler et al.*, 2016], but farther downwind (within hours)
- other species dominate [Dunlea et al., 2007; Ninneman et al., 2019]; Schwab et al., 2009].
- 85 Although nitric acid vapor and partially oxidized alkyl nitrates may stick to inlets, the loss may
- be reversible and other species such as PAN and simple alkyl nitrates pass through sample lines
- 87 like NO₂. Nighttime chemistry can also be important for air and water quality ozone reacts
- 88 with NO₂ to yield NO₃, N₂O₅, and related species thus NO₉ can greatly exceed NOx even in
- 89 winter [Brown and Stutz, 2012; Brown et al., 2012]. Most AQS-reported NOx is therefore more
- 90 accurately considered NOy, with NOy HNO₃ as a lower limit. Evaluating model NOx
- 91 concentrations with output from monitors with heated converters will lead to erroneous
- 92 conclusions for winter chemistry as well. Because of the variety of interfering species, the

93 correction to measurements from commercial NOx instruments with hot molybdenum varies

94 with the hydrocarbon mix as well as photochemical environment, and thus with time of day and

season. The overestimate in NOx has led the EPA to call for "true NOx" in the Enhanced

96 Monitoring Plans (80 FR 65292; October 26, 2015).

97

98 Using NOx monitor data for research-grade studies is challenging, and methods vary widely. An 99 interference correction algorithm for monitor results presented in the AQS has been developed 100 for comparison of remotely sensed NO₂ to surface sites. Ordonez et al. (2006) compared measurements from the Global Ozone Monitoring Experiment (GOME) spectrometer to ground based 101 102 in situ measurements. They reported much better agreement with vertical column NO₂ when interferences were taken into account. The ratio of true NO₂ to that measured with conventional 103 commercial analyzers ranged from 0.858 in January to 0.485 in June [Ordonez et al., 2006]. To 104 105 infer ground level NO₂ concentrations from the Ozone Monitoring Instrument (OMI) a correction algorithm was developed to estimate the conversion factor, CF: 106

107

108

$$CF = \frac{NO_2}{NO_2 + \Sigma \ N + \ 0.95P \ N) + 0.35HNO_3)}$$

109

110 [Lamsal et al., 2008]. This has sometimes been applied when comparing measurements and models [Souri et al., 2017; Souri et al., 2018]; although it is strictly only valid for the satellite 111 overpass time and generally clear skies, the method appears to give reasonable results when 112 appropriate uncertainty is considered. These corrections may not be applicable in winter or at 113 114 night when N₂O₅ chemistry can be active and important. Determining trends in NOx can rely on 115 consistent measurements even if interferences are substantial. For example, Kang et al. (2013) 116 used NOx from AQS to investigate trends, but limited results to only 6-9am locally when the boundary layer is shallow and vehicular emissions are fresh, although the ratio of NOx to NOy 117 118 falls with distance from major sources, and could cause a bias. The authors describe the NOx analysis as a qualitative measure, and they appear to successfully distinguish changes in 119 emissions vs. meteorology and conclude that emissions as model input are inconsistent with 120 121 observed changes. In an study of trends over the US, investigators tried to minimize the 122 interferences by focusing on early morning measurements when the model indicated NOy is 123 composed mostly of NOx [Tong et al., 2015]; these methods appear to be adequate to distinguish 124 trends in emissions during the global recession. 125

126 Other studies appear to compare modeled NOx directly to measurements with a heated converter.

127 In Beijing, the role of regional transport in an ozone episode was recently determined with the

- Community Multiscale Air Quality (CMAQ) [*Liu et al.*, 2019]. Commercial NOx analyzers with
 sensitivity to NOz species
- 130 (http://www.mee.gov.cn/gkml/sthjbgw/stbgth/201809/W020180905375349444950.pdf) were
- $\label{eq:concentrations} 131 \qquad \text{employed to evaluate modeled NOx. Although NO}_2 \text{ concentrations were generally high, often}$
- above 100 μ g/m³ (~50 ppb), interfering species could contribute substantially to measurements
- in more rural locations such as Shanxi Province.
- 134
- 135 In a five-city study [*Friberg et al.*, 2017] CMAQ simulations of air pollution were compared to
- measurements of a variety of species including NOx. Data from the State and Local Air
- 137 Monitoring Stations (SLAMS) network were used. As reported by the USEPA

- 138 (https://www3.epa.gov/ttn/amtic/files/nearroad/NearRoadTAD.pdf) these instruments are almost
- exclusively commercial NOx analyzers with heated converters. It is inappropriate to compared
- 140 modeled NOx to output from monitors with known interferences. The interfering species could
- 141 contribute to the low spatiotemporal correlations observed for NOx relative to high values seen
- 142 for secondary species such as ozone. Comparison of modeled NOy to monitor output or use of a
- 143 CF would be more appropriate and might lead to better agreement.
- 144
- 145 To obtain air pollution data at fine resolution, a model fusion approach was applied and
- evaluated with observations from monitors in the Central Speciation Network (CSN) and
- 147 Southeast Aerosol Research and Characterization network (SEARCH) network as well as
- 148 monitors operated by the Georgia Environmental Protection Department [*Bates et al.*, 2018].
- 149 While the SEARCH monitors employ photolytic conversion of NO₂ to NO [*Hansen et al.*, 2003]
- and are thus more nearly specific to NOx, many monitors employed for this study were
- 151 commercial instruments with interferences
- 152 (https://airgeorgia.org/informationaboutno2.html#NO3). Modeled NOx should not be compared
- to these measurements. This inherent sensitivity to additional species in commercial NOx
- analyzers could generate small errors for near-road monitors and major bias in monitors farther
- 155 from sources; these errors could compromise epidemiological studies.
- 156
- 157 A recent paper [*Qin et al.*, 2019] investigates the sensitivity of ozone in CMAQ to input
- variables including emissions. The authors compared modeled NOx to AQS-reported
- 159 measurements in the Great Lakes area (interferences are not discussed) and concluded that
- 160 "using CB6 combined with a 30% reduction of on-road mobile NOx emissions and MEGAN led
- to the best performance." They indicate that this result is superior to runs with a 50% NOx
- 162 emissions reduction. But commercial monitors do not allow for such precision; modeled NOx
- 163 was apparently compared to instruments that more nearly measured NOy. The model domain
- 164 includes much of the eastern United States where on average NOy greatly exceeds NOx (see
- above); emissions overestimates are likely substantially larger than those reported.
- 166
- 167 Quantitative assessment of ozone formation and effective policy require not just reasonable168 simulation of ozone and relative reduction factors, but also good estimates of ozone precursors
- 169 especially NOx and NOy. To be reliable, models must get ozone right for the right reasons.
- 170 Several modeling studies found substantially better agreement [*Canty et al.*, 2015; *Mao et al.*,
- 171 2018] when a 50% or greater reduction was applied to vehicular NOx emissions. Errors in NO
- 172 concentration impact the fate of HO_2 and RO_2 and cascade throughout the entire simulation. We
- 173 suggest that model results provide quantitative guidance only when emissions are evaluated
- against reliable measurements; these results add urgency to the deployment of true NO_2 and true
- 175 NOy monitors. Until such data are available, modeled NOy (minus some fraction of HNO₃) is
- 176 more comparable to measurements with heated converters.
- 177
- Comparing model NOx with measurements from commercial monitors can lead to fundamental
 errors NOx concentrations impact radicals and alter the relative role of NOx vs. VOCs in the
- 180 rate of ozone production. More realistic comparison of simulations to measurements may well
- indicate substantially greater model high bias for reactive nitrogen and better simulate the
- 182 response of the ozone in the atmosphere to controls on NOx emissions. This would also bring
- 183 model results into better agreement with research indicating a historical overestimate of NOx

184 emissions in the National Emissions Inventory (NEI), for example [Anderson et al., 2014;

185 Brioude et al., 2013; Castellanos et al., 2011; McDonald et al., 2018; Travis et al., 2016].

186 Policy relevant science requires careful evaluation of observations and appropriate comparison to

- simulations.
- 188

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193 **References**

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